Classical and quantum controllability of a rotating 3D symmetric molecule

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Abstract

In this paper we study the controllability problem for a symmetric-top molecule, both for its classical and quantum rotational dynamics. As controlled fields we consider three orthogonally polarized electric fields which interact with the electric dipole of the molecule. We characterize the controllability in terms of the dipole position: when it lies along the symmetry axis of the molecule neither the classical nor the quantum dynamics are controllable, due to the presence of a conserved quantity, the third component of the total angular momentum; when it lies in the orthogonal plane to the symmetry axis, a quantum symmetry arises, due to the superposition of symmetric states, which has no classical counterpart. If the dipole is neither along the symmetry axis nor orthogonal to it, controllability for the classical dynamics and approximate controllability for the quantum dynamics are proved to hold. The controllability properties of the classical rotational dynamics are analyzed by applying geometric control theory techniques. To establish the approximate controllability of the symmetric-top Schrödinger equation we use a Lie-Galerkin method, based on block-wise approximations of the infinite dimensional systems.

Key words: Quantum control, Schrödinger equation, rotational dynamics, symmetric molecule, bilinear control systems

AMS Classification: 68Q25, 68R10,68U05

Introduction

The control of molecular dynamics takes an important role in quantum physics and chemistry because of the variety of its applications, starting from well-established ones such as rotational state-selective excitation of chiral molecules ([15, 14]), and going further to applications in quantum information ([25]). For a general overview of controlled molecular dynamics one can see, for example, [19].

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Rotations can, in general, couple to vibrations in the so-called ro-vibrational states. In our mathematical analysis, however, we shall restrict ourselves to the rotational states of the molecule. Due to its discrete quantization, molecular dynamics perfectly fits the mathematical quantum control theory which has been established until now. In fact, the control of the Schrödinger equation has attracted substantial interest in the last 15 years. See, for instance, [8, 7, 10, 9, 11, 13, 21, 5, 2, 18] for the state of the art.

The problem of controlling the rotational dynamics of a planar molecule has been analyzed in [7] by means of two orthogonally polarized electric fields; in particular, approximate controllability has been proved using non-resonant connectedness chains in the spectrum of the rotational Hamiltonian. In [8] the approximate controllability of a linear-top controlled by three orthogonally polarized electric fields has been established. There, a new sufficient condition for controllability, called the Lie–Galerkin tracking condition, has been introduced in an abstract framework, and applied to the linear-top system.

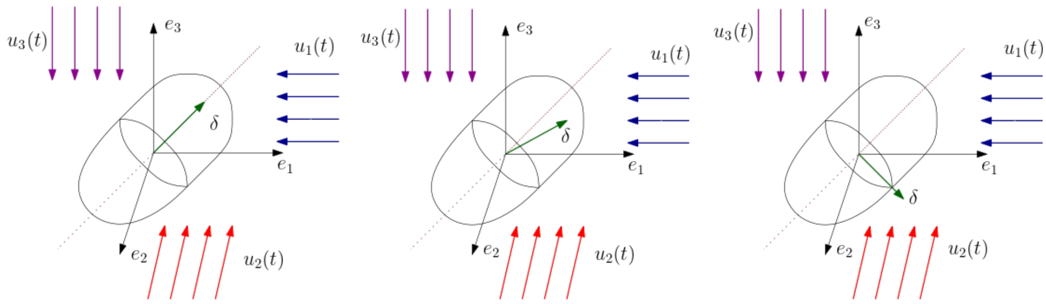
Here, we study the symmetric-top as a generalization of the linear one, characterizing its controllability in terms of the position of its dipole moment in Theorems 6, 7, 9. While for the linear-top two quantum numbers j, m are needed to describe the motion, the main and more evident difference here is the presence of a third quantum number k, which classically represents the projection of the total angular momentum on the symmetry axis of the molecule. This should not be a surprise, since the configuration space of a linear-top is the 2-sphere S^2, while the symmetric-top evolves in the Lie group $\mathrm{SO}(3)$, a three-dimensional manifold. As a matter of fact, by fixing $k = 0$, one recovers the linear-top as a subsystem inside the symmetric-top. It is worth mentioning that the general theory developed in [11, 7, 21] is based on non-resonance conditions on the spectrum of the internal Hamiltonian. A major difficulty in studying the controllability properties of the rotational dynamics is that, even in the case of the linear-top, the spectrum of the rotational Hamiltonian has severe degeneracies at the so-called m-levels. The symmetric-top is even more degenerate, due to the additional presence of the so-called k-levels.

The Schrödinger equation for a rotating molecule controlled by three orthogonally polarized electric fields reads

$$\mathrm{i}\frac{\partial}{\partial t}\psi(R,t) = H\psi(R,t) + \sum_{l=1}^{3} u_l(t)B_l(R,\delta)\psi(R,t), \quad \psi(\cdot,t) \in L^2(\mathrm{SO}(3)),$$

where $H = \frac{1}{2}\Big(\frac{P_1^2}{I_1} + \frac{P_2^2}{I_2} + \frac{P_3^2}{I_3}\Big)$ is the rotational Hamiltonian, I_1, I_2, I_3 are the moments of inertia of the molecule, P_1, P_2, P_3 are the angular momentum differential operators, and $B_i(R,\delta) = -\langle R\delta, e_i\rangle$ is the interaction Hamiltonian between the dipole moment δ of the molecule and the direction e_i, $i = 1, 2, 3$. Finally, $R \in \mathrm{SO}(3)$ is the matrix which describes the configuration of the molecule in the space.

We shall study the symmetric-top, that is, the case $I_1 = I_2$. In this case, closed expression for the spectrum and the eigenfunctions of H are known. The case of the asymmetric-top, which goes beyond the scope of this paper, could be tackled either by a perturbative approach or with a further computational effort. Here, the position of the dipole moment plays a decisive role: when it is neither along the symmetry axis, nor orthogonal to it, as in the above Figure (b), then approximate controllability holds, under some non-resonance conditions, as it is stated in Theorem 7. To prove it, we introduce in Section 2.1 a block-wise Lie–Galerkin condition, closely related to the Lie–Galerkin tracking condition, which is shown to provide a general controllability test for the multi-input Schrödinger equation (Theorem 5). We then apply this result to the symmetric-top system. The control strategy is based on the excitation of the system with external fields in resonance with three families of frequencies, corresponding to internal



(a) (b) (c)

Figure 1:

spectral gaps. One frequency is used to overcome the m-degeneracy in the spectrum, and this step is quite similar to the proof of the linear-top's approximate controllability (Appendix A). The other two frequencies are used in a next step to break the k-degeneracy, in a three-wave mixing scheme (Appendix B) typically used in quantum chemistry to obtain enantio- and state-selectivity for chiral molecules ([16],[24],[3]).

The two dipole configurations to which the Theorem 7 does not apply are extremely relevant from the physical point of view. Indeed, the dipole moment of a symmetric-top lies usually along its symmetry axis (Figure (a)), and if not, for accidentally symmetric-top, it is often found in the orthogonal plane (Figure (c)). Here two different symmetries arise, implying the non-controllability of these systems, as we prove, respectively, in Theorems 6 and 9. These two conserved quantities stimulated and motivated the study of the classical dynamics of the symmetric-top, presented in the first part of the paper: the first conserved quantity, appearing in Theorem 6, corresponds to a classical observable, that is, the component of the angular momentum along the symmetry axis, and it turns out to be a prime integral also for the classical controlled dynamics, as remarked in Theorem 2. The second conserved quantity, appearing in Theorem 9, is more challenging, because it does not have a counterpart in the classical dynamics, being mainly due to the superposition of k and $-k$ states in the quantum dynamics. We show that this position of the dipole still correspond to a controllable system for the classical-top, while it does not for the quantum-top. Thus, the latter is an example of a system whose quantum dynamics are not controllable even though the classical dynamics are. The possible discrepancy between quantum and classical controllability has been already noticed, for example, in the harmonic oscillator dynamics ([20]). It should be noticed that the classical controllability of a rigid body has been analyzed in several works like [1, Section 6.4], [17, Section 4.6] using internal controls; nevertheless here we adopt as control fields three external forces, a strategy which has never been considered, as far as we know, in the previous literature.

The paper is organized as follows: in Section 1 we study the controllability of the classical Hamilton equations for a symmetric-top. The main results are summarized in Theorems 2 and 3, where we prove, respectively, the non-controllability when the dipole lies along the symmetry axis of the body and the controllability in any other case. In Section 2 we study the controllability of the Schrödinger equation for a symmetric-top. The main controllability result is Theorem 7, where we prove the approximate controllability when the dipole is neither along the symmetry axis, nor orthogonal to it. In the two cases left, we prove the non-controllability in Theorems 6 and 9.

1 Classical symmetric-top molecule

1.1 Controllability of control-affine systems with recurrent drift

We recall in this section some useful results on the controllability properties of (finite-dimensional) control-affine systems.

Let M be an n-dimensional manifold, $X_0, X_1, \dots, X_m$ a family of smooth (i.e., C^∞) vector fields on M, $U \subset \mathbb{R}^m$ a set of control values which contains a neighbourhood of the origin. We consider the controlled system

$$\dot{q} = X_0(q) + \sum_{i=1}^{m} u_i(t) X_i(q), \qquad q \in M, \tag{1}$$

where the control functions u are taken in $L^\infty(\mathbb{R}, U)$. The vector field X_0 is called the *drift*.

- The *reachable set* from $q_0 \in M$ is

$$\begin{aligned}\mathrm{Reach}(q_0) := &\{q \in M \mid \exists\, u, T \text{ s.t. the solution to (1) with } q(0) = q_0 \\ &\text{satisfies } q(T) = q\}.\end{aligned}$$

- The system (1) is said to be *controllable* if $\mathrm{Reach}(q_0) = M$ for all $q_0 \in M$.
- The family of vector fields $X_0, X_1, \dots, X_m$ is said to be *Lie bracket generating* if

$$\dim(\mathrm{Lie}_q\{X_0, X_1, \dots, X_m\}) = n$$

  for all $q \in M$, where $\mathrm{Lie}_q\{X_0, X_1, \dots, X_m\}$ denotes the evaluation of q of the Lie algebra generated by $X_0, X_1, \dots, X_m$.
- Let X be a complete vector field on M and ϕ^t its flow at time t, $t \in \mathbb{R}$. Then X is said to be *recurrent* if for every open nonempty subset V of M and every time $t > 0$, there exists $\tilde{t} > t$ such that $\phi^{\tilde{t}}(V) \cap V \neq \emptyset$.

The following is a basic result in geometric control theory (see, for example, [17, Section 4.6]).

Theorem 1. *If X_0 is recurrent and the family $X_0, X_1, \dots, X_m$ is Lie bracket generating, then system (1) is controllable.*

Using the *Orbit Theorem* (see, e.g., [1, Chapter 5]) and assuming the analyticity of the vector fields, one proves the following result, which is useful to verify the Lie bracket generating condition.

Lemma 1. *If the family of analytic vector fields $X_0, X_1, \dots, X_m$ is Lie bracket generating on the complement of a sub-manifold $N \subset M$ and $\mathrm{Reach}(q) \not\subset N$, for all $q \in N$, then the family is Lie bracket generating on M.*

1.2 Hamilton equations on a Lie group

The dynamics on a Lie group G, relative to a Hamiltonian function H on $T^*G \cong G \times \mathfrak{g}^*$, where $\mathfrak{g}$ is the Lie algebra of G, can be described in terms of the Hamiltonian field $\vec{H}$, defined as a vector field on T^*G by $dH(V) = \omega(\vec{H}, V)$, $V \in T(T^*G)$, where ω is the canonical symplectic 2-form on

T^*G. We briefly describe this classical approach to dynamical systems on Lie groups (see, for instance, [17, Chapter 12]). First we identify

$$T(T^*G) \cong T(G \times \mathfrak{g}^*) \cong TG \times T\mathfrak{g}^* \cong (G \times \mathfrak{g}) \times (\mathfrak{g}^* \times \mathfrak{g}^*),$$

that is, an element of $T(T^*G)$ can be seen as $((g, X), (p, Y^*))$ where (X, Y^*) is a tangent vector with base (g, p) and $X(g, p) \in \mathfrak{g}$, $Y^*(g, p) \in \mathfrak{g}^*$, for all $(g, p) \in G \times \mathfrak{g}^*$.

If V is a vector field on T^*G, we write $V = (X, Y^*)$ as before, and f is a function on T^*G, V acts as a derivation on f by the formula

$$(Vf)(g, p) = \Big((L_g)^* \frac{\partial f}{\partial g}(g, p)\Big) X(g, p) + Y^*(g, p)\Big(\frac{\partial f}{\partial p}(g, p)\Big)$$

for all $(g, p) \in G \times \mathfrak{g}^*$, where $\dfrac{\partial f}{\partial g} \in T_g^*G$, $\dfrac{\partial f}{\partial p} \in T_p^*\mathfrak{g}^* \cong \mathfrak{g}^{**} \cong \mathfrak{g}$.

Notice that $V_1 = (X_1, Y_1^*)$, $V_2 = (X_2, Y_2^*)$ are tangent vectors at (q, p), then we have $\omega(V_1, V_2) = Y_2^*(X_1) - Y_1^*(X_2) - ((\mathrm{ad}X_1)^*p)X_2$. As a consequence the expression of the Hamiltonian field $\vec{H} = (X, Y^*)$ in terms of its defining Hamiltonian function H is

$$\begin{cases} X(g, p) = \dfrac{\partial H}{\partial p}(g, p) \\ Y^*(g, p) = -(L_g)^*\Big(\dfrac{\partial H}{\partial g}(g, p)\Big) - (adX)^*p. \end{cases}$$

Thus the integral curves $t \mapsto (g(t), p(t)) \in G \times \mathfrak{g}^*$ of $\vec{H}$ satisfy

$$\begin{cases} \dfrac{dg(t)}{dt} = (L_{g(t)})_* \dfrac{\partial H}{\partial p}(g(t), p(t)) \\ \dfrac{dp(t)}{dt} = -(L_{g(t)})^*\Big(\dfrac{\partial H}{\partial g}(g(t), p(t))\Big) - \Big(ad\dfrac{\partial H}{\partial p}(g(t), p(t))\Big)^* p(t). \end{cases} \tag{2}$$

These are the equations of motion on G.

Remark 1. *If G is abelian, then* $\Big(ad\dfrac{\partial H}{\partial p}\Big)^* = 0$. *Thus one recovers the classical Hamiltonian equations. If H is left-invariant, then* $\dfrac{\partial H}{\partial g} = 0$. *Thus one obtains the so-called equations in vertical coordinates.*

1.3 The classical dynamics of a molecule subject to electric fields

Since the molecule is a rigid body that can just rotate, the configuration space is the Lie group SO(3). We now apply the machinery just developed for a general Lie group to $G = \mathrm{SO}(3)$. Consider the basis

$$\widehat{A_1} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -1 \\ 0 & 1 & 0 \end{pmatrix}, \qquad \widehat{A_2} = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ -1 & 0 & 0 \end{pmatrix}, \qquad \widehat{A_3} = \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},$$

of $\mathfrak{g} = \mathfrak{so}(3)$. Let $\mathbf{A_1}, \mathbf{A_2}, \mathbf{A_3}$ be the corresponding left-invariant vector fields. Denoting by e_1, e_2, e_3 a fixed orthonormal frame on $\mathbb{R}^3$ and by a_1, a_2, a_3 a fixed orthonormal frame on the rigid body with the same orientation, the configuration of the molecule is identified with the unique $g \in \mathrm{SO}(3)$ such that $ga_i = e_i$, for $i = 1, 2, 3$.

As external forces on the system we consider the three electric fields with intensities $u_1(t), u_2(t), u_3(t)$ and directions e_1, e_2, e_3. Denoting by δ the dipole of the molecule written in the moving frame, the three potential energies due to the interaction with the electric fields are

$$V_i = -u_i(t)\langle g\delta, e_i\rangle, \qquad i = 1, 2, 3.$$

Set $V := \sum_{i=1}^{3} V_i$ the total potential energy. Finally let H_i, for $i = 1, 2, 3$, be the (left-invariant) Hamiltonian function relative to the left-invariant vector field $\mathbf{A_i}$ (that is, $H_i(p) := p(\widehat{A_i}), \forall p \in \mathfrak{so}(3)^*$) and we introduce the Hamiltonian of the dynamical system

$$H := \frac{1}{2}\Big(\frac{H_1^2}{I_1} + \frac{H_2^2}{I_2} + \frac{H_3^2}{I_3}\Big) + V, \tag{3}$$

defined on $\mathrm{SO}(3) \times \mathfrak{so}(3)^*$, where I_1, I_2, I_3 are the moments of inertia of the rigid body in a moving frame made by principal axes of inertia.

Remark 2. *H is not left-invariant since $V = V(g)$.*

Applying (2), the integral curves $(g(t), p(t)) \in \mathrm{SO}(3) \times \mathfrak{so}(3)^*$ of $\vec{H}$ satisfy

$$\begin{cases} \dfrac{dg(t)}{dt} = \sum_{i=1}^{3} \dfrac{1}{I_i} H_i(p(t))\mathbf{A_i}(g(t)), \\ \dfrac{dp(t)}{dt} = -(L_{g(t)})^*(dV) - (\mathrm{ad}\Omega(p(t)))^* p(t), \end{cases} \tag{4}$$

where $\Omega(p) := \sum_{i=1}^{3} \frac{1}{I_i} H_i(p)\widehat{A_i}$.

Now using the isomorphism $\mathfrak{so}(3)^* \to \mathfrak{so}(3)$, $p \mapsto \widehat{P}$, defined by $K(\widehat{P}, \widehat{A}) = p(\widehat{A})$, where $K(\cdot,\cdot)$ is the scalar product on $\mathfrak{so}(3)$ given by $K(\widehat{A}, \widehat{B}) = -\frac{1}{2}\mathrm{tr}(\widehat{A}\widehat{B}), \forall \widehat{A}, \widehat{B} \in \mathfrak{so}(3)$, and the classical identification of Lie algebras

$$(\mathfrak{so}(3), [\cdot,\cdot]) \to (\mathbb{R}^3, \times), \quad \widehat{A} = \begin{pmatrix} 0 & -a_3 & a_2 \\ a_3 & 0 & -a_1 \\ -a_2 & a_1 & 0 \end{pmatrix} \mapsto A = \begin{pmatrix} a_1 \\ a_2 \\ a_3 \end{pmatrix},$$

where $\times$ is the vector product, the Hamilton equations (4) read on $\mathrm{SO}(3) \times \mathbb{R}^3$

$$\begin{cases} \dfrac{dg(t)}{dt} = g(t)\widehat{\Omega}(t), \\ \dfrac{dP(t)}{dt} = P(t) \times \Omega(t) + \sum_{i=1}^{3} u_i(t)(g^{-1}(t)e_i) \times \delta. \end{cases} \tag{5}$$

These are the Euler equations for a charged rigid body subject to three orthogonal electric fields. We shall also use the notation $S(A)$ for the anti-symmetric matrix $\widehat{A}$.

System (5) can be seen as a control-affine system with three controlled fields: by computing

$$\Omega(t) = \Omega(p(t)) = \sum_{i=1}^{3} \frac{1}{I_i} H_i(p(t))A_i = \sum_{i=1}^{3} \frac{1}{I_i} p(t)(\widehat{A_i})A_i = \sum_{i=1}^{3} \frac{1}{I_i} P_i(t)A_i,$$

we obtain $\Omega(p(t)) = \beta P(t)$, where $\beta P = (P_1/I_2, P_2/I_2, P_3/I_3)^t$. Then (5) reads

$$\begin{pmatrix} \dot{g} \\ \dot{P} \end{pmatrix} = X(g, P) + \sum_{i=1}^{3} u_i(t)Y_i(g, P), \quad (g, P) \in \mathrm{SO}(3) \times \mathbb{R}^3, \tag{6}$$

where

$$X(g,P) := \begin{pmatrix} gS(\beta P) \\ P \times (\beta P) \end{pmatrix}, \quad Y_i(g,P) := \begin{pmatrix} 0 \\ (g^{-1}e_i) \times \delta \end{pmatrix}, \quad i = 1,2,3. \tag{7}$$

Rotating molecule dynamics can also be represented in terms of quaternions. The 3-sphere S^3 (seen as a double covering space of $\mathrm{SO}(3)$) inherits a Lie group structure from the quaternions $\mathbb{H}$ and its elements act as rotations on $\mathbb{R}^3$. Let us now reformulate Euler equations (5) in this formalism.

We identify $S^3 = \{q_0 + \mathrm{i}q_1 + \mathrm{j}q_2 + \mathrm{k}q_3 \mid (q_0,q_1,q_2,q_3) \in \mathbb{R}^4, q_0^2+q_1^2+q_2^2+q_3^2 = 1\} \subset \mathbb{H}$ and $T_1S^3 = \mathbb{R}^3 = \{\mathrm{i}P_1 + \mathrm{j}P_2 + \mathrm{k}P_3 \mid (0,P_1,P_2,P_3) \in \mathbb{R}^4\} \subset \mathbb{H}$. Thus, if $q \in S^3$ and $P \in \mathbb{R}^3$, q rotates P by the formula $qP\overline{q}$ as a quaternions multiplication. Via this identification, the vector product $P \times \Omega$ becames $\frac{1}{2}[P,\Omega] := \frac{1}{2}(P\Omega - \Omega P)$. Thus, we get

$$\begin{cases} \dfrac{dq(t)}{dt} = & q(t)\Omega(t), \\ \dfrac{dP(t)}{dt} = & \dfrac{1}{2}[P(t),\Omega(t)] + \dfrac{u_1(t)}{2}[\overline{q(t)}\mathrm{i}q(t),\delta] + \dfrac{u_2(t)}{2}[\overline{q(t)}\mathrm{j}q(t),\delta] \\ & + \dfrac{u_3(t)}{2}[\overline{q(t)}\mathrm{k}q(t),\delta]. \end{cases} \tag{8}$$

This description will be useful for computations.

1.4 Non-controllability of the classical genuine symmetric-top

In most cases of physical interest, the electric dipole δ of a symmetric-top molecule lies along the symmetry axis of the molecule. If $I_1 = I_2$, the symmetry axis is the third one, and we have that $\delta = \begin{pmatrix} 0 \\ 0 \\ \delta_3 \end{pmatrix}$, $\delta_3 \neq 0$, in the body frame. The corresponding molecule is called a *genuine symmetric-top*.

Theorem 2. *The third angular momentum P_3 is a conserved quantity for the controlled motion* (6) *of the genuine symmetry-top molecule.*

Proof. In order to compute the equation satisfied by P_3 in (6), notice that

$$P(t) \times \beta P(t) = \begin{pmatrix} P_1(t) \\ P_2(t) \\ P_3(t) \end{pmatrix} \times \begin{pmatrix} P_1(t)/I_2 \\ P_2(t)/I_2 \\ P_3(t)/I_3 \end{pmatrix} = \begin{pmatrix} \left(\frac{1}{I_3} - \frac{1}{I_2}\right)P_2(t)P_3(t) \\ \left(\frac{1}{I_2} - \frac{1}{I_3}\right)P_1(t)P_3(t) \\ 0 \end{pmatrix}.$$

Moreover, $u_i(t)(g^{-1}(t)e_i) \times \delta = u_i(t)(g^{-1}(t)e_i) \times \begin{pmatrix} 0 \\ 0 \\ \delta_3 \end{pmatrix} = \begin{pmatrix} \star \\ \star \\ 0 \end{pmatrix}$. Hence, for a genuine symmetric-top, the equation for P_3 becomes $\dfrac{dP_3(t)}{dt} = 0$. □

As a consequence, the controlled dynamics live in the hypersurfaces $\{P_3 = \mathrm{const}\}$ and hence system (6) is not controllable in the 6-dimensional manifold $\mathrm{SO}(3) \times \mathbb{R}^3$.

1.5 Controllability of the classical accidentally symmetric-top

In Theorem 2 we proved that P_3 is a prime integral for equations (5), using both the symmetry of the mass and the symmetry of the charge, meaning that $I_1 = I_2$ and $\delta = (0, 0, \delta_3)^t$. We consider now a symmetric-top molecule with electric dipole δ not along the symmetry axis of the body, that is, $\delta = \begin{pmatrix} \delta_1 \\ \delta_2 \\ \delta_3 \end{pmatrix}$, with $\delta_1 \neq 0$ or $\delta_2 \neq 0$. This system is usually called *accidentally symmetric-top*.

Theorem 3. *For an accidentally symmetric-top molecule system* (6) *is controllable.*

Proof. The drift X is recurrent, as observed in [1, Section 8.4]. Thus, by Theorem 1, to prove controllability it suffices to show that, for any $(g, P) \in \mathrm{SO}(3) \times \mathbb{R}^3$, $\dim\Big(\mathrm{Lie}_{(g,P)}\{X, Y_1, Y_2, Y_3\}\Big) = 6$. Actually, we will find six vector fields on $\mathrm{SO}(3) \times \mathbb{R}^3$ such that their span is six-dimensional everywhere but on a hypersurface, and we will conclude by applying Lemma 1. Notice that, since

$$[DY_i]Y_j = \begin{pmatrix} 0 & 0 \\ \star & 0 \end{pmatrix} \begin{pmatrix} 0 \\ (g^{-1}e_j) \times \delta \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix},$$

the control vector fields commute: $[Y_i, Y_j] = 0$, for $i, j = 1, 2, 3$. Moreover, $[X, Y_i](g, P) = \begin{pmatrix} -gS(\beta[(g^{-1})e_i \times \delta]) \\ \star \end{pmatrix}$. Denote by $\Pi_{\mathrm{SO}(3)}$ the projection onto the $\mathrm{SO}(3)$ part of the tangent bundle, that is,

$$\Pi_{\mathrm{SO}(3)} : T_{(g,P)}(\mathrm{SO}(3) \times \mathbb{R}^3) = T_g\mathrm{SO}(3) \times T_P\mathbb{R}^3 \to T_g\mathrm{SO}(3).$$

Then we have

$$\begin{aligned} &\mathrm{span}\{\Pi_{\mathrm{SO}(3)}X(g,P), \Pi_{\mathrm{SO}(3)}[X, Y_1](g,P), \Pi_{\mathrm{SO}(3)}[X, Y_2](g,P), \Pi_{\mathrm{SO}(3)}[X, Y_3](g,P)\} \\ &\quad = gS\Big(\beta[\{\delta\}^\perp \oplus \mathrm{span}\{P\}]\Big). \end{aligned}$$

Hence, if $\langle P, \delta \rangle \neq 0$, we have

$$\begin{aligned} \dim\Big(&\mathrm{span}\{\Pi_{\mathrm{SO}(3)}X(g,P), \Pi_{\mathrm{SO}(3)}[X, Y_1](g,P), \Pi_{\mathrm{SO}(3)}[X, Y_2](g,P), \\ &\Pi_{\mathrm{SO}(3)}[X, Y_3](g,P)\}\Big) = 3. \end{aligned} \tag{9}$$

To go further in the analysis, it is convenient to use the quaternionic parametrization (8) in which every field is polynomial. We have, in coordinates $q = (q_0, q_1, q_2, q_3) \in S^3, P = (P_1, P_2, P_3) \in \mathbb{R}^3$,

$$X(q, P) = \begin{pmatrix} q\beta P \\ \frac{1}{2}[P, \beta P] \end{pmatrix} = \begin{pmatrix} -q_1\frac{P_1}{I_2} - q_2\frac{P_2}{I_2} - q_3\frac{P_3}{I_3} \\ q_0\frac{P_1}{I_2} + q_2\frac{P_3}{I_3} - q_3\frac{P_2}{I_2} \\ q_0\frac{P_2}{I_2} - q_1\frac{P_3}{I_3} + q_3\frac{P_1}{I_2} \\ q_0\frac{P_3}{I_3} + q_1\frac{P_2}{I_2} - q_2\frac{P_1}{I_2} \\ \left(\frac{1}{I_3} - \frac{1}{I_2}\right)P_2P_3 \\ \left(\frac{1}{I_2} - \frac{1}{I_3}\right)P_1P_3 \\ 0 \end{pmatrix},$$

$$
Y_1(q,P) = \begin{pmatrix} 0 \\ \frac{1}{2}[\overline{q}\mathrm{i}q, \delta] \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \\ (q_1q_2 - q_0q_3)\delta_3 - (q_1q_3 + q_0q_2)\delta_2 \\ (q_1q_3 + q_0q_2)\delta_1 - \frac{1}{2}(q_0^2 + q_1^2 - q_2^2 - q_3^2)\delta_3 \\ \frac{1}{2}(q_0^2 + q_1^2 - q_2^2 - q_3^2)\delta_2 - (q_1q_2 - q_0q_3)\delta_1 \end{pmatrix},
$$

$$
Y_2(q,P) = \begin{pmatrix} 0 \\ \frac{1}{2}[\overline{q}\mathrm{j}q, \delta] \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \\ \frac{1}{2}(q_0^2 - q_1^2 + q_2^2 - q_3^2)\delta_3 - (q_2q_3 - q_0q_1)\delta_2 \\ (q_2q_3 - q_0q_1)\delta_1 - (q_1q_2 + q_0q_3)\delta_3 \\ (q_1q_2 + q_0q_3)\delta_2 - \frac{1}{2}(q_0^2 - q_1^2 + q_2^2 - q_3^2)\delta_1 \end{pmatrix}.
$$

Let us consider the six vector fields $X, Y_1, Y_2, [X, Y_1], [X, Y_2], [[X, Y_1], Y_1]$: we have that a 6×6 minor of the matrix

$$
(X(q,P), Y_1(q,P), Y_2(q,P), [X, Y_1](q,P), [X, Y_2](q,P), [[X, Y_1], Y_1](q,P))
$$

has determinant equal to $S(q,P) := S_1(q)S_2(q)S_3(q)S_4(q)S_5(P)$, where

$$
\begin{aligned}
S_1(q) &:= \frac{I_2 - I_3}{I_2^2 I_3^2} q_1, \\
S_2(q) &:= (-2q_1q_2\delta_1 + 2q_0q_3\delta_1 + q_0^2\delta_2 + q_1^2\delta_2 - (q_2^2 + q_3^2)\delta_2), \\
S_3(q) &:= (q_0(-2q_2\delta_1 + 2q_1\delta_2) + 2q_3(q_1\delta_1 + q_2\delta_2) + (q_0^2 - q_1^2 - q_2^2 + q_3^2)\delta_3)^2, \\
S_4(q) &:= (-2(q_0q_2 + q_1q_3)(\delta_1^2 + \delta_2^2) + ((q_0^2 + q_1^2 - q_2^2 - q_3^2)\delta_1 + 2(q_1q_2 - q_0q_3)\delta_2)\delta_3), \\
S_5(P) &:= P_1\delta_1 + P_2\delta_2 + P_3\delta_3 = \langle P, \delta \rangle.
\end{aligned}
$$

Hence, for all (q,P) such that $S(q,P) \neq 0$,

$$
\begin{aligned}
\dim \Big(\mathrm{span}\{ & X(q,P), Y_1(q,P), Y_2(q,P), [X, Y_1](q,P), [X, Y_2](q,P), \\
& [[X, Y_1], Y_1](q,P)\} \Big) = 6,
\end{aligned}
$$

that is, outside the hypersurface $V(S) := \{(q,P) \in S^3 \times \mathbb{R}^3 \mid S(q,P) = 0\}$ the family X, Y_1, Y_2 is Lie bracket generating.

Now we are left to prove that

$$
\mathrm{Reach}(q,P) \not\subset V(S), \quad \forall (q,P) \in V(S),
$$

and then to apply Lemma 1. Let us start by considering the factor S_5 of S and notice that, for any fixed $q \in S^3$, $V(S_5)$ defines a surface inside $\mathbb{R}^3$. Denote by $\Pi_{\mathbb{R}^3}$ the projection onto the $\mathbb{R}^3$ part of the tangent bundle, that is,

$$
\Pi_{\mathbb{R}^3} : T_{(q,P)}(S^3 \times \mathbb{R}^3) = T_qS^3 \times T_P\mathbb{R}^3 \to T_P\mathbb{R}^3 = \mathbb{R}^3.
$$

The vector field $\Pi_{\mathbb{R}^3} X$ is tangent to $V(S_5)$ when

$$\langle \nabla_P S_5, \Pi_{\mathbb{R}^3} X \rangle = \langle \delta, [P, \beta P] \rangle = 0,$$

that is, if and only if $P_3 = 0$ or $P_2\delta_1 - P_1\delta_2 = 0$. Notice that one vector between $\Pi_{\mathbb{R}^3} Y_1, \Pi_{\mathbb{R}^3} Y_2, \Pi_{\mathbb{R}^3} Y_3$ is not tangent to $V(P_3)$, otherwise

$$\operatorname{span}\{\Pi_{\mathbb{R}^3} Y_1, \Pi_{\mathbb{R}^3} Y_2, \Pi_{\mathbb{R}^3} Y_3\} = \{P_3 = 0\} = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}^{\perp}.$$

However,

$$\operatorname{span}\{\Pi_{\mathbb{R}^3} Y_1, \Pi_{\mathbb{R}^3} Y_2, \Pi_{\mathbb{R}^3} Y_3\} = \delta^{\perp},$$

which would imply that δ is collinear to $(0,0,1)^t$, which is impossible since the molecule is accidentally symmetric.

Concerning the hypersurface $V(P_2\delta_1 - P_1\delta_2)$, we consider again $\Pi_{\mathbb{R}^3} X$, which is tangent when $\langle \nabla_P (P_2\delta_1 - P_1\delta_2), \Pi_{\mathbb{R}^3} X \rangle = 0$, that is, if and only if $P_3 = 0$ or $P_1\delta_1 + P_2\delta_2 = 0$. We treat the second case, being $P_3 = 0$ already treated. Hence, we consider the intersection

$$\begin{cases} P_2\delta_1 - P_1\delta_2 = 0, \\ P_1\delta_1 + P_2\delta_2 = 0. \end{cases}$$

The only solution of the system is $P_1 = P_2 = 0$, because the molecule is accidentally symmetric. Finally, when $P_1 = P_2 = 0$, we consider the two-dimensional distribution $\operatorname{span}\{\Pi_{\mathbb{R}^3} Y_1, \Pi_{\mathbb{R}^3} Y_2, \Pi_{\mathbb{R}^3} Y_3\}$, which cannot be tangent to the P_3 axis.

Summarizing, if δ is not collinear to $(0,0,1)^t$, we have

$$\operatorname{Reach}(q, P) \not\subset V(S_5), \quad \forall (q, P) \in V(S_5).$$

To conclude, if $(q, P) \in V(S_i)$, $i = 1, \ldots, 4$, then we fix P and we get a two-dimensional manifold $\{q \in S^3 \mid S_i(q) = 0\} \subset S^3$. Now the projections of the vector fields $X, [X, Y_1], [X, Y_2], [X, Y_3]$ on the base part of the bundle span a three-dimensional vector space if $\langle P, \delta \rangle \neq 0$, as observed in (9). So, by possibly steering P until $\langle P, \delta \rangle \neq 0$, it is possible to exit from $V(S_i)$. This concludes the proof of the theorem. □

1.6 Reachable sets of the classical genuine symmetric-top

Theorem 2 states that the hypersurfaces $\{P_3 = \text{const}\}$ are invariant for the controlled motion. Next we prove that the restriction of system (5) to any such hypersurface is controllable.

Theorem 4. *Let $I_1 = I_2$ and $\delta = (0, 0, \delta_3)^t$, $\delta_3 \neq 0$. Then for $(\overline{g}, \overline{P}) \in \mathrm{SO}(3) \times \mathbb{R}^3$, $\overline{P} = (\overline{P_1}, \overline{P_2}, \overline{P_3})$, one has*

$$\operatorname{Reach}(\overline{g}, \overline{P}) = \{(g, P) \in \mathrm{SO}(3) \times \mathbb{R}^3 \mid P_3 = \overline{P_3}\}.$$

Proof. From Theorem 2 we know that $\{P_3 = \text{const}\}$ is invariant. Since the drift X is recurrent, it suffices to prove that system (5) is Lie bracket generating on the 5-dimensional manifold $\{P_3 = \text{const}\}$.

We recall from (9) that, if $\langle P, \delta \rangle \neq 0$, that is, if $P_3 \neq 0$, we have

$$\dim\Big(\operatorname{span}\{\Pi_{\mathrm{SO}(3)} X(g, P), \Pi_{\mathrm{SO}(3)}[X, Y_1](g, P), \Pi_{\mathrm{SO}(3)}[X, Y_2](g, P), \Pi_{\mathrm{SO}(3)}[X, Y_3](g, P)\}\Big) = 3.$$

Moreover, taking the projections of the control fields on the $\mathbb{R}^3$ part of the bundle $\Pi_{\mathbb{R}^3} Y_i(q,P) = (g^{-1}e_i)\times\delta$ we see that

$$\dim\Big(\operatorname{span}\{\Pi_{\mathbb{R}^3}Y_1,\Pi_{\mathbb{R}^3}Y_2,\Pi_{\mathbb{R}^3}Y_3\}\Big) = 2 \tag{10}$$

everywhere. Thus, if $P_3 \neq 0$, we have that

$$\dim\Big(\operatorname{span}\{X(g,P),Y_1(g,P),Y_2(g,P),Y_3(g,P),[X,Y_1](g,P),[X,Y_2](g,P),\\ [X,Y_3](g,P)\}\Big) = 5.$$

So the system is Lie bracket generating on the manifold $\{P_3 = \text{const} \neq 0\}$.

We are left to consider the case $P_3 = 0$. Notice that $\Pi_{\mathbb{R}^3}Y_1,\Pi_{\mathbb{R}^3}Y_2,\Pi_{\mathbb{R}^3}Y_3$ give a two-dimensional distribution for any value of P_3. So we consider in the quaternionic parametrization the projections of $X,[X,Y_1],[X,Y_2],[[X,Y_1],X]$ on the S^3 part of the bundle and we obtain

$$\dim\Big(\operatorname{span}\{\Pi_{S^3}X(q,P),\Pi_{S^3}[X,Y_1](q,P),\Pi_{S^3}[X,Y_2](q,P),\\ \Pi_{S^3}[[X,Y_1],X](q,P)\}\Big) = 3,$$

for $P_3 = 0$, except when $q_3[2P_2(q_1q_2 - q_0q_3) + P_1(q_0^2+q_1^2-q_2^2-q_3^2)] = 0$. This equation defines the union of two surfaces inside S^3. (Notice that we can assume $P_1 \neq 0$ and $P_2 \neq 0$ because (10) gives local controllability in (P_1,P_2)). On $\{q_3 = 0\}$, we have that $\Pi_{S^3}X$ is tangent if and only if $q_1P_2 - q_2P_1 = 0$. On the curve $\gamma \subset S^3$ of equation

$$\begin{cases} q_3 = 0, \\ q_1P_2 - q_2P_1 = 0, \end{cases}$$

we can consider the two-dimensional distribution spanned by $\Pi_{S^3}[X,Y_1],\Pi_{S^3}[X,Y_2],\Pi_{S^3}[X,Y_3]$, which is clearly not tangent to γ. Following Lemma 1, the system is Lie bracket generating also on $\{q_3 = 0\}$.

Analogously, on $\{2P_2(q_1q_2 - q_0q_3) + P_1(q_0^2+q_1^2-q_2^2-q_3^2) = 0\}$ we consider the vector field $\Pi_{S^3}[[[X,Y_1],X],Y_2]$ which is tangent if and only if $(q_0q_2+q_1q_3)(P_1q_0q_1 + P_2q_0q_2 - P_2q_1q_3 + P_1q_2q_3) = 0$. Again, since the distribution spanned by $\Pi_{S^3}[X,Y_1],\Pi_{S^3}[X,Y_2],\Pi_{S^3}[X,Y_3]$ is two-dimensional, we can exit from the set of equation

$$\begin{cases} 2P_2(q_1q_2 - q_0q_3) + P_1(q_0^2+q_1^2-q_2^2-q_3^2) = 0, \\ (q_0q_2+q_1q_3)(P_1q_0q_1 + P_2q_0q_2 - P_2q_1q_3 + P_1q_2q_3) = 0, \end{cases}$$

whose strata have dimension at most one. Thus, applying again Lemma 1, we can conclude that the restriction of the system on the manifold $\{P_3 = 0\}$ is Lie bracket generating. $\square$

2 Quantum symmetric-top molecule

2.1 Controllability of the multi-input Schrödinger equation

Let $\ell \in \mathbb{N}$, $a > 0$ and $U := [-a,a]^\ell$. Let $\mathcal{H}$ be an infinite-dimensional Hilbert space with scalar product $\langle\cdot,\cdot\rangle$ (linear in the first entry and conjugate linear in the second), $H, B_1, \dots, B_\ell$ be (possibly unbounded) self-adjoint operators on $\mathcal{H}$, with domains $D(H), D(B_1), \ldots, D(B_\ell)$. We consider the controlled Schrödinger equation

$$\mathrm{i}\frac{d\psi(t)}{dt} = (H + \sum_{j=1}^{\ell} u_j(t)B_j)\psi(t), \quad \psi(t) \in \mathcal{H}, \quad u(t) \in U. \tag{11}$$

Definition 1. • *We say that the operator H satisfies ($\mathbb{A}1$) if it has discrete spectrum with infinitely many distinct eigenvalues (possibly degenerate).*
Denote by $\mathcal{B}$ a Hilbert basis $(\phi_k)_{k\in\mathbb{N}}$ of $\mathcal{H}$ made of eigenvectors of H associated with the family of eigenvalues $(\lambda_k)_{k\in\mathbb{N}}$ and let $\mathcal{L}$ be the set of finite linear combination of eigenstates, that is,

$$\mathcal{L} = \mathrm{span}\{\phi_k \mid k \in \mathbb{N}\}.$$

- *We say that $(H, B_1, \ldots, B_\ell, \mathcal{B})$ satisfies ($\mathbb{A}2$) if $\phi_k \in D(B_j)$ for every $k \in \mathbb{N}$, $j = 1, \ldots, \ell$.*
- *We say that $(H, B_1, \ldots, B_\ell, \mathcal{B})$ satisfies ($\mathbb{A}3$) if*

$$H + \sum_{j=1}^{\ell} u_j B_j : \mathcal{L} \to \mathcal{H}$$

is essentially self-adjoint for every $u \in U$.

- *We say that $(H, B_1, \ldots, B_\ell, \mathcal{B})$ satisfies ($\mathbb{A}$) if H satisfies ($\mathbb{A}1$) and $(H, B_1, \ldots, B_\ell, \mathcal{B})$ satisfies ($\mathbb{A}2$) and ($\mathbb{A}3$).*

If $(H, B_1, \ldots, B_\ell, \mathcal{B})$ satisfies ($\mathbb{A}$) then, for every $(u_1, \ldots, u_\ell) \in U$, $H + \sum_{j=1}^{\ell} u_j B_j$ generates a subgroup $e^{-\mathrm{i}t(H+\sum_{j=1}^{\ell} u_j B_j)}$ of the group of unitary operators $U(\mathcal{H})$. It is therefore possible to define the propagator Γ_T^u at time T of system (11) associated with a piecewise constant control law $u(\cdot) = (u_1(\cdot), \ldots, u_\ell(\cdot))$ by composition of flows of the type $e^{-\mathrm{i}t(H+\sum_{j=1}^{\ell} u_j B_j)}$. If, moreover, $B_1, \ldots, B_\ell$ are bounded operators then the definition can be extended by continuity to every L^∞ control law (see [4]).

Definition 2. *Let $(H, B_1, \ldots, B_\ell, \mathcal{B})$ satisfy ($\mathbb{A}$).*

- *Given ψ_0, ψ_1 in the unit sphere $\mathcal{S}$ of $\mathcal{H}$, we say that ψ_1 is reachable from ψ_0 if there exist a time $T > 0$ and a piecewise constant control law $u : [0, T] \to U$ such that $\psi_1 = \Gamma_T^u(\psi_0)$. We denote by $\mathrm{Reach}(\psi_0)$ the set of reachable points from ψ_0.*
- *We say that (11) is approximately controllable if for every $\psi_0 \in \mathcal{S}$ the set $\mathrm{Reach}(\psi_0)$ is dense in $\mathcal{S}$.*

We introduce the notion of module-tracker (m-tracker, for brevity) that is, a system for which any given curve can be tracked up to (relative) phases. The identification up to phases of elements of $\mathcal{H}$ in the basis $\mathcal{B} = (\phi_k)_{k\in\mathbb{N}}$ can be accomplished by the projection

$$\mathcal{M} : \psi \mapsto \sum_{k\in\mathbb{N}} |\langle \phi_k, \psi\rangle| \phi_k.$$

Definition 3. *Let $(H, B_1, \ldots, B_\ell, \mathcal{B})$ satisfy ($\mathbb{A}$). We say that system (11) is an* m-tracker *if, for every $r \in \mathbb{N}$, $\psi_1, \ldots, \psi_r$ in $\mathcal{H}$, $\widehat{\Gamma} : [0, T] \to U(\mathcal{H})$ continuous with $\widehat{\Gamma}_0 = \mathrm{Id}_{\mathcal{H}}$, and $\epsilon > 0$, there exists an invertible increasing continuous function $\tau : [0, T] \to [0, T_\tau]$ and a piecewise constant control $u : [0, T_\tau] \to U$ such that*

$$\|\mathcal{M}(\widehat{\Gamma}_t \psi_k) - \mathcal{M}(\Gamma^u_{\tau(t)} \psi_k)\| < \epsilon, \qquad k = 1, \ldots, r,$$

for every $t \in [0, T_\tau]$.

Remark 3. *We recall that if system* (11) *is an m-tracker, then it is also approximately controllable, as noticed in [8, Remark 2.9].*

We now introduce some objects in order to state a new sufficient condition for a system to be m-tracker, based on the controllability results in [8].

Let $\{I_j\}_{j\in\mathbb{N}}$ be a family of subsets of $\mathbb{N}$ such that

$$\cup_{j\in\mathbb{N}} I_j = \mathbb{N} \quad \text{and} \quad n_j := |I_j| < \infty, \forall j \in \mathbb{N},$$

where $|I_j|$ denotes the cardinality of I_j. Consider the subspaces

$$\mathcal{M}_j := \operatorname{span}\{\phi_n \mid n \in I_j\} \subset \mathcal{H}.$$

Let us denote by $\Sigma_j := \{|\lambda_l - \lambda_{l'}| \mid l, l' \in I_j\}$ the spectral gaps in $\mathcal{M}_j$, and define the orthogonal projections

$$\Pi_{\mathcal{M}_j} : \mathcal{H} \ni \psi \mapsto \sum_{n\in I_j} \langle \phi_n, \psi\rangle \phi_n \in \mathcal{H}.$$

Given a linear operator Q on $\mathcal{H}$ we identify the linear operator $\Pi_{\mathcal{M}_j} Q \Pi_{\mathcal{M}_j}$ preserving $\mathcal{M}_j$ with its complex matrix representations with respect to the basis $(\phi_n)_{n\in I_j}$. We define $B_i^{(j)} := \Pi_{\mathcal{M}_j} B_i \Pi_{\mathcal{M}_j}$ for every $i = 1, \ldots, \ell$.

We define

$$\Xi_j^0 = \{(\sigma, i) \in \Sigma_j \times \{1, \ldots, \ell\} \mid (B_i)_{l,k} = 0 \text{ for every } l \in \mathbb{N},\ k \in \mathbb{N} \setminus I_j \\ \text{such that } |\lambda_l - \lambda_k| = \sigma\},$$

and

$$\Xi_j^1 = \{(\sigma, i) \in \Sigma_j \times \{1, \ldots, \ell\} \mid (B_i)_{l,k} = 0 \text{ for every } l \in I_j,\ k \in \mathbb{N} \setminus I_j \\ \text{such that } |\lambda_l - \lambda_k| = \sigma\}.$$

While the set Ξ_j^0 is made by totally non-resonant gaps, in Ξ_j^1 the resonances corresponding to pairs of eigenstates outside I_j are allowed.

For every $\sigma \geq 0$, and every square matrix M of dimension m, let

$$\mathcal{E}_\sigma(M) = (M_{l,k} \delta_{\sigma, |\lambda_l - \lambda_k|})_{l,k=1,\ldots,m},$$

where $\delta_{l,k}$ is the Kronecker delta. The square matrix $\mathcal{E}_\sigma(B_i^{(j)})$ of dimension n_j, $i = 1, \ldots, \ell$, corresponds to the activation in $B_i^{(j)}$ of the spectral gap $\sigma \in \Sigma_j$: every element is 0 except the (l,k)-elements such that $|\lambda_l - \lambda_k| = \sigma$. Moreover, for every $\xi \in S^1 \subset \mathbb{C}$, we consider the matrix operator W_ξ such that

$$(W_\xi(M))_{l,k} = \begin{cases} \xi M_{l,k}, & \lambda_l < \lambda_k, \\ 0, & \lambda_l = \lambda_k, \\ \bar{\xi} M_{l,k}, & \lambda_l > \lambda_k. \end{cases} \tag{12}$$

Next, we consider the sets of excited modes

$$\nu_j^s := \{W_\xi(\mathcal{E}_\sigma(\mathrm{i}B_i^{(j)})) \mid (\sigma, i) \in \Xi_j^s, \sigma \neq 0, \xi \in S^1\}, \quad s = 0, 1. \tag{13}$$

Notice that $\nu_j^0 \subset \nu_j^1 \subset \mathfrak{su}(n_j)$. We denote by $\operatorname{Lie}(\nu_j^s)$ the Lie algebra generated by the matrices in ν_j^s, $s = 0, 1$, with respect to the bracket of $\mathfrak{su}(n_j)$, and define $\mathcal{T}_j$ as the minimal ideal of $\operatorname{Lie}(\nu_j^1)$ containing ν_j^0.

Finally, we introduce the graph $\mathcal{G}$ with vertices $\mathcal{V} = \{I_j\}_{j\in\mathbb{N}}$ and edges $\mathcal{E} = \{(I_j, I_k) \mid j, k \in \mathbb{N},\ I_j \cap I_k \neq \emptyset\}$. We are now in a position to state a sufficient condition for a system to be an m-tracker, and thus, approximately controllable.

Theorem 5. *Assume that* ($\mathbb{A}$) *holds true. If the graph $\mathcal{G}$ is connected and $\mathcal{T}_j = \mathfrak{su}(n_j)$, for every $j \in \mathbb{N}$, then* (11) *is an m-tracker.*

Proof. We shall prove that the assumptions of the theorem imply that system (11) satisfies the Lie–Galerkin tracking condition ([8, Definition 2.7]) and hence that (11) is an m-tracker ([8, Theorem 2.8]).

Up to reordering the sets I_j, we can assume that

$$I_{j+1} \cap (\cup_{k=1}^{j} I_k) \neq \emptyset, \qquad \forall j \in \mathbb{N}. \tag{14}$$

Let us fix $n_0 \in \mathbb{N}$ and consider $m \in \mathbb{N}$ such that $\{\phi_1, \ldots, \phi_{n_0}\} \subset \sum_{j=1}^{m} \mathcal{M}_j$. Then, according to [8, Definition 2.7], we shall prove that

$$\mathrm{Lie}(\cup_{j=1}^{m} \widetilde{\mathcal{T}}_j) = \mathfrak{su}(\dim(\sum_{j=1}^{m} \mathcal{M}_j)), \tag{15}$$

where the set of operators $\widetilde{\mathcal{T}}_j$ is obtained similarly to $\mathcal{T}_j$, replacing the set of operators ν_j^0, ν_j^1 by

$$\{W_\xi(\mathcal{E}_\sigma(\mathrm{i}\Pi_{\sum_{j=1}^{m} \mathcal{M}_j} B_i \Pi_{\sum_{j=1}^{m} \mathcal{M}_j})) \mid (\sigma, i) \in \Xi_j^s, \sigma \neq 0, \xi \in S^1\}, \quad s = 0, 1.$$

We proceed by induction on m. For $m = 1$, (15) is true, since we have that $\mathrm{Lie}(\mathcal{T}_1) = \mathcal{T}_1 = \mathfrak{su}(n_1) = \mathfrak{su}(\dim(\mathcal{M}_1))$. Assume now that (15) is true for m, and consider the vertex I_{m+1}. Consider $t, p \in \cup_{j=1}^{m+1} I_j$ and let us prove that $E_{t,p} := e_{t,p} - e_{p,t}$ is in $\mathrm{Lie}(\cup_{j=1}^{m+1} \widetilde{\mathcal{T}}_j)$, where $e_{a,b}$ is the matrix with entries all 0 except for the one in row a and column b, which is equal to 1 (and the indices in $\cup_{j=1}^{m+1} I_j$ are identified with the elements of $\{1, \ldots, \dim(\sum_{j=1}^{m+1} \mathcal{M}_j)\}$). Notice that, by definition of Ξ_j^0, Ξ_j^1, a matrix in $\widetilde{\mathcal{T}}_{m+1}$ has the form

$$\left[\begin{array}{c|c} 0 & 0 \\ \hline 0 & Q \end{array}\right], \qquad Q \in \mathfrak{su}(n_{m+1}),$$

where the upper-left square matrix has dimension $\dim(\sum_{j=1}^{m+1} \mathcal{M}_j) - n_{m+1}$. Similarly, a matrix in $\cup_{j=1}^{m} \widetilde{\mathcal{T}}_j$ has the form

$$\left[\begin{array}{c|c} Q & 0 \\ \hline 0 & 0 \end{array}\right], \qquad Q \in \mathfrak{su}(\dim(\sum_{j=1}^{m} \mathcal{M}_j)),$$

where the lower-right square matrix has dimension $\dim(\sum_{j=1}^{m+1} \mathcal{M}_j) - \dim(\sum_{j=1}^{m} \mathcal{M}_j)$.

If $t, p \in \cup_{j=1}^{m} I_j$ or $t, p \in I_{m+1}$ the conclusion follows from the induction hypothesis and the identity $\mathcal{T}_{m+1} = \mathfrak{su}(n_{m+1})$. Let then $t \in I_{m+1} \setminus (\cup_{j=1}^{m} I_j)$ and $p \in \cup_{j=1}^{m} I_j$. Fix, moreover, $r \in I_{m+1} \cap (\cup_{j=1}^{m} I_j)$, whose existence is guaranteed by (14). Again by the induction hypothesis and the identity $\mathcal{T}_{m+1} = \mathfrak{su}(n_{m+1})$, we have that $E_{p,r}$ and $E_{r,t}$ are in $\mathrm{Lie}(\cup_{j=1}^{m+1} \widetilde{\mathcal{T}}_j)$. The bracket $[E_{p,r}, E_{r,t}] = E_{p,t}$ is therefore also in $\mathrm{Lie}(\cup_{j=1}^{m+1} \widetilde{\mathcal{T}}_j)$. By similar arguments, we deduce that every basis element of $\mathfrak{su}(\dim(\sum_{j=1}^{m+1} \mathcal{M}_j))$ is in $\mathrm{Lie}(\cup_{j=1}^{m+1} \widetilde{\mathcal{T}}_j)$. □

2.2 The Schrödinger equation of a symmetric-top subject to electric fields

We recall in this section some general facts about Wigner functions and the theory of angular momentum in quantum mechanics (see, for instance, [6, 23, 12]).

We use Euler's angles $(\alpha, \beta, \gamma) \in [0, 2\pi) \times [0, \pi] \times [0, 2\pi)$ to describe the configuration space $\mathrm{SO}(3)$ of the molecule. More precisely, the coordinates of a vector change from the body fixed frame a_1, a_2, a_3 to the space fixed frame e_1, e_2, e_3 via three rotations

$$\begin{pmatrix} X \\ Y \\ Z \end{pmatrix} = R_{e_3}(\alpha) R_{e_2}(\beta) R_{e_3}(\gamma) \begin{pmatrix} x \\ y \\ z \end{pmatrix} =: R(\alpha, \beta, \gamma) \begin{pmatrix} x \\ y \\ z \end{pmatrix} \tag{16}$$

where $(x, y, z)^t$ are the coordinates of the vector in the body fixed frame, $(X, Y, Z)^t$ are the coordinates of the vector in the space fixed frame and $R_{e_i}(\theta) \in \mathrm{SO}(3)$ is a rotation of angle θ around the axis e_i. The explicit expression of the matrix $R(\alpha, \beta, \gamma) \in \mathrm{SO}(3)$ is

$$R = \begin{pmatrix} \cos\alpha\cos\beta\cos\gamma - \sin\alpha\sin\gamma & -\cos\alpha\cos\beta\sin\gamma - \sin\alpha\cos\gamma & \cos\alpha\sin\beta \\ \sin\alpha\cos\beta\cos\gamma + \cos\alpha\sin\gamma & -\sin\alpha\cos\beta\sin\gamma + \cos\alpha\cos\gamma & \sin\alpha\sin\beta \\ -\sin\beta\cos\gamma & \sin\beta\sin\gamma & \cos\beta \end{pmatrix}. \tag{17}$$

In Euler coordinates, the angular momentum operators are given by

$$\begin{cases} J_1 = \mathrm{i}\cos\alpha\cot\beta\dfrac{\partial}{\partial\alpha} + \mathrm{i}\sin\alpha\dfrac{\partial}{\partial\beta} - \mathrm{i}\dfrac{\cos\alpha}{\sin\beta}\dfrac{\partial}{\partial\gamma}, \\ J_2 = \mathrm{i}\sin\alpha\cot\beta\dfrac{\partial}{\partial\alpha} - \mathrm{i}\cos\alpha\dfrac{\partial}{\partial\beta} - \mathrm{i}\dfrac{\sin\alpha}{\sin\beta}\dfrac{\partial}{\partial\gamma}, \\ J_3 = -\mathrm{i}\dfrac{\partial}{\partial\alpha}. \end{cases} \tag{18}$$

These are linear operators acting on the Hilbert space $L^2(\mathrm{SO}(3))$, self-adjoint with respect to the Haar measure $\frac{1}{8}d\alpha d\gamma \sin\beta d\beta$. Using (18), the self-adjoint operator $P_3 := -\mathrm{i}\frac{\partial}{\partial\gamma}$ can be written as $P_3 = \sin\beta\cos\alpha J_1 + \sin\beta\sin\alpha J_2 + \cos\beta J_3$, that is,

$$P_3 = \sum_{i=1}^{3} R_{i3}(\alpha, \beta, \gamma) J_i,$$

where $R = (R_{ij})_{i,j=1}^3$ is given in (17).

In the same way we define $P_1 = \sum_{i=1}^3 R_{i1}(\alpha, \beta, \gamma) J_i$, $P_2 = \sum_{i=1}^3 R_{i2}(\alpha, \beta, \gamma) J_i$. The operators J_i and P_i, $i = 1, 2, 3$, are the angular momentum operators expressed in the fixed and in the body frame, respectively. Finally, we consider the square norm operator $J^2 := J_1^2 + J_2^2 + J_3^2 = P_1^2 + P_2^2 + P_3^2$. Now, J^2, J_3, P_3 can be considered as the three commuting observables needed to describe the quantum motion of a molecule. Indeed, $[J^2, J_3] = [J^2, P_3] = [J_3, P_3] = 0$, and hence there exists an orthonormal Hilbert basis of $L^2(\mathrm{SO}(3))$ which diagonalizes simultaneously J^2, J_3 and P_3. In terms of Euler coordinates, this basis is made by the so-called Wigner functions ([23, Chapter 4], [6, Section 3.8,3.9])

$$D^j_{k,m}(\alpha, \beta, \gamma) := e^{\mathrm{i}(m\alpha + k\gamma)} d^j_{k,m}(\beta), \qquad j \in \mathbb{N}, \quad k, m = -j, \dots, j, \tag{19}$$

where

$$d^j_{k,m}(\beta) := n_{j,k,m} \sin\Big(\frac{\beta}{2}\Big)^{|k-m|} \cos\Big(\frac{\beta}{2}\Big)^{|k+m|} F\Big(\sin\Big(\frac{\beta}{2}\Big)^2\Big).$$

The function $F\Big(\sin\Big(\frac{\beta}{2}\Big)^2\Big)$ is an hypergeometric series and $n_{j,k,m}$ is a normalizing factor.

Summarizing, the family of Wigner functions $\{D^j_{k,m} \mid j \in \mathbb{N}, k, m = -j, \dots, j\}$ forms an orthonormal Hilbert basis for $L^2(\mathrm{SO}(3))$. Moreover,

$$\begin{cases} J^2 D^j_{k,m} = j(j+1) D^j_{k,m}, \\ J_3 D^j_{k,m} = m D^j_{k,m}, \\ P_3 D^j_{k,m} = k D^j_{k,m}. \end{cases}$$

Thus, m and k are the quantum numbers which correspond respectively to the projections of the angular momentum on the third axis of the fixed and the body frame.

Remark 4. *The level $k = 0$ corresponds to the vanishing of the third angle γ. Then the system becomes a linear molecule which can be described by the spherical harmonics $\{Y^j_m \mid j \in \mathbb{N}, m = -j, \dots, j\}$ ([23, Chapter 5], [6, Section 3.10]). Indeed, $Y^j_m(\alpha, \beta) = D^j_{0,m}(\alpha, \beta, \gamma)$.*

The rotational Hamiltonian of a molecule is $H = \frac{1}{2}\Big(\frac{P_1^2}{I_1} + \frac{P_2^2}{I_2} + \frac{P_3^2}{I_3}\Big)$, which is seen here as a self-adjoint operator acting on the Hilbert space $L^2(\mathrm{SO}(3))$. From now on, we impose the symmetry condition for the molecule, that is, $I_1 = I_2$, which implies that $H = \frac{J^2}{2I_2} + \Big(\frac{1}{2I_3} - \frac{1}{2I_2}\Big)P_3^2$. Thus,

$$H D^j_{k,m} = \Big(\frac{j(j+1)}{2I_2} + \Big(\frac{1}{2I_3} - \frac{1}{2I_2}\Big)k^2\Big) D^j_{k,m} =: E^j_k D^j_{k,m}. \tag{20}$$

Hence, the Wigner functions are the eigenfunctions of H, i.e., the rotational states of the symmetric top and its eigenvalues E^j_k, $j \in \mathbb{N}, k = -j, \dots, j$, are the rotational energies of the molecule. Since the eigenvalues of H do not depend on m, the energy level E^j_k is $(2j+1)$-degenerate with respect to m. This property is common to every molecule in nature: the spectrum $\sigma(H)$ does not depend on m, just like in classical mechanics kinetic energy does not depend on the direction of the angular momentum. Moreover, the energy level E^j_k is also 2-degenerate with respect to k, when $k \neq 0$. This extra degeneracy is actually a characterizing property of every symmetric molecule, which is not verified by asymmetric ones. Breaking this k-symmetry will be one important feature of our controllability analysis.

The interaction Hamiltonian between the dipole δ inside the molecule and the external electric field in the direction e_i, $i = 1, 2, 3$, is given by the Stark effect ([12, Chapter 10])

$$B_i(\alpha, \beta, \gamma) = -\langle R(\alpha, \beta, \gamma)\delta, e_i \rangle,$$

seen as a multiplicative self-adjoint operator acting on $L^2(\mathrm{SO}(3))$. Thus, the rotational Schrödinger equation for a symmetric-top molecule subject to three orthogonally polarized electric fields reads

$$\mathrm{i}\frac{\partial}{\partial t}\psi(\alpha, \beta, \gamma; t) = H\psi(\alpha, \beta, \gamma; t) + \sum_{l=1}^{3} u_l(t) B_l(\alpha, \beta, \gamma)\psi(\alpha, \beta, \gamma; t), \quad \psi(t) \in L^2(\mathrm{SO}(3)), \tag{21}$$

$u(t) \in [-a, a]^3, a > 0$.

2.3 Non-controllability of the quantum genuine symmetric-top

We recall that the genuine symmetric-top molecule is a symmetric rigid body with electric dipole δ along the symmetry axis: $\delta = \begin{pmatrix} 0 \\ 0 \\ \delta_3 \end{pmatrix}$ in the principal axis frame on the body. We then introduce the subspaces $S_k := \overline{\text{span}}\{D^j_{k,m} \mid j \in \mathbb{N}, m = -j, \dots, j\}$, where $\overline{\text{span}}$ denotes the closure of the linear hull in $L^2(\mathrm{SO}(3))$.

Theorem 6. *The quantum number k is invariant in the controlled motion of the genuine symmetric-top molecule. That is, if $I_1 = I_2$ and $\delta = \begin{pmatrix} 0 \\ 0 \\ \delta_3 \end{pmatrix}$, the subspaces S_k are invariant for any propagator of the Schrödinger equation (21).*

Proof. We have to show that H and B_l, for $l = 1, 2, 3$, do not couple different levels of k, that is,

$$\begin{cases} \langle D^j_{k,m}, \mathrm{i}HD^{j'}_{k',m'} \rangle_{L^2(\mathrm{SO}(3))} = 0, \quad k \neq k', \\ \langle D^j_{k,m}, \mathrm{i}B_l D^{j'}_{k',m'} \rangle_{L^2(\mathrm{SO}(3))} = 0, \quad k \neq k', \ l = 1, 2, 3. \end{cases} \tag{22}$$

The first equation of (22) is obvious since the orthonormal basis $\{D^j_{k,m}\}$ diagonalizes H. Under the genuine symmetric-top assumption, the second equation of (22) is also true: for $l = 1$ and $k \neq k'$ we compute

$$\begin{aligned} &\langle D^j_{k,m}, \mathrm{i}B_1 D^{j'}_{k',m'} \rangle_{L^2(\mathrm{SO}(3))} \\ &= -\int_0^{2\pi} d\alpha \int_0^{2\pi} d\gamma \int_0^{\pi} d\beta \sin(\beta) D^j_{k,m}(\alpha,\beta,\gamma) \mathrm{i}B_1(\alpha,\beta,\gamma) \overline{D^{j'}_{k',m'}}(\alpha,\beta,\gamma) \\ &= -\mathrm{i}\delta_3 \Big(\int_0^{2\pi} d\gamma e^{\mathrm{i}k\gamma} e^{-\mathrm{i}k'\gamma} \Big) \\ &\Big(\int_0^{2\pi} d\alpha \int_0^{\pi} d\beta (-\sin^2(\beta)\cos(\alpha) e^{\mathrm{i}m\alpha} e^{-\mathrm{i}m'\alpha} d^j_{k,m}(\beta) \overline{d^{j'}_{k',m'}}(\beta) \Big) = 0, \end{aligned}$$

using the orthogonality of the functions $e^{ik\gamma}$ and the explicit form (17) of the matrix R, which yields

$$B_1(\alpha,\beta,\gamma) = -\langle \begin{pmatrix} 0 \\ 0 \\ \delta_3 \end{pmatrix}, R^{-1}(\alpha,\beta,\gamma) \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} \rangle = -\delta_3 \sin\beta \cos\alpha.$$

The computations for $l = 2, 3$ are analogous, since the multiplicative potentials B_l do not depend on γ. □

Remark 5. *Equation (22) also shows that, for a genuine symmetric-top, the third component of the angular momentum P_3 commutes with B_l, i.e.,*

$$[P_3, B_l] = 0, \quad l = 1, 2, 3.$$

Thus, by Noether Theorem ([22, Theorem 8.1]), we have that $\langle \psi(t), P_3\psi(t) \rangle$ is a conserved quantity, where ψ is the solution of (21).

2.4 Controllability of the quantum accidentally symmetric-top

So far we have studied the dynamics of a symmetric-top molecule with electric dipole moment along its symmetry axis and we have proven that its dynamics are trapped in the eigenspaces of P_3.

Nevertheless, for applications to molecules charged in the laboratory, or to particular molecules present in nature such as D_2S_2 (Figure 2.6) or H_2S_2, it is interesting to consider also the case in which the dipole has non-zero components outside the symmetry axis: this case is called the *accidentally symmetric molecule*.

Under a non-resonance condition, we are going to prove that, if the dipole moment is not orthogonal to the symmetry axis of the molecule, the rotational dynamics of an accidentally symmetric-top are approximately controllable. To prove this statement, we are going to show that the hypothesis of Theorem 5 are satisfied by (21).

Theorem 7. *Let $I_1 = I_2$, $\frac{I_2}{I_3} \notin \mathbb{Q}$. If $\delta = (\delta_1, \delta_2, \delta_3)^t$ is such that $\delta \neq \begin{pmatrix} 0 \\ 0 \\ \delta_3 \end{pmatrix}$ and $\delta \neq \begin{pmatrix} \delta_1 \\ \delta_2 \\ 0 \end{pmatrix}$, then the Schrödinger equation (21) is an m-tracker.*

Proof. First of all, one can check, for example in [12, Table 2.1], that the pairings induced by the interaction Hamiltonians satisfy

$$\langle D^j_{k,m}, \mathrm{i}B_l D^{j'}_{k',m'} \rangle = 0, \tag{23}$$

when $|j' - j| > 1$, or $|k' - k| > 1$ or $|m' - m| > 1$, for every $l = 1, 2, 3$. Equation (23) is the general form of the so-called selection rules.

We then define for every $j \in \mathbb{N}$ the set $I_j := \{\rho(l, k, m) \mid l = j, j+1,\ k, m = -l, \dots, l\} \subset \mathbb{N}$, where $\rho : \{(l, k, m) \mid l \in \mathbb{N}, k, m = -l, \dots, l\} \to \mathbb{N}$ is the lexicographic ordering. The graph $\mathcal{G}$ whose vertices are the sets I_j and whose edges are $\{(I_j, I_{j'}) \mid I_j \cap I_{j'} \neq \emptyset\} = \{(I_j, I_{j+1}) \mid j \in \mathbb{N}\}$ is therefore linear. In order to apply Theorem 5 we shall then consider the projection of (21) onto each space $\mathcal{M}_j := \mathcal{H}_j \oplus \mathcal{H}_{j+1}$, where $\mathcal{H}_l := \mathrm{span}\{D^l_{k,m} \mid k, m = -l, \dots, l\}$. The dimension of $\mathcal{M}_j$ is $(2j+1)^2 + (2(j+1)+1)^2$, and we identify $\mathfrak{su}(\mathcal{M}_j)$ with $\mathfrak{su}((2j+1)^2 + (2(j+1)+1)^2)$.

According to (23), the three types of spectral gaps in $\mathcal{M}_j$ which we should consider are

$$\lambda^j_k := |E^{j+1}_{k+1} - E^j_k| = \left| \frac{j+1}{I_2} + \Big(\frac{1}{2I_3} - \frac{1}{2I_2} \Big)(2k+1) \right|, \quad k = -j, \dots, j, \tag{24}$$

corresponding to pairings for which both j and k move (see Figure 2),

$$\eta_k := |E^j_{k+1} - E^j_k| = \left| \Big(\frac{1}{2I_3} - \frac{1}{2I_2} \Big)(2k+1) \right|, \quad k = -j, \dots, j, \tag{25}$$

and

$$\sigma^j := |E^{j+1}_k - E^j_k| = \frac{j+1}{I_2}, \tag{26}$$

for which, respectively, only k or j moves (see, Figures 3(a) and 3(b)).

Having introduced the spectral gaps needed in our analysis, we now classify them with respect to the sets Ξ^0_j and Ξ^1_j introduced in Section 2.1.

Lemma 2 (External resonances). *Let $I_2/I_3 \notin \mathbb{Q}$. Then $(\lambda^j_k, l), (\sigma^j, l) \in \Xi^0_j$, and $(\eta_k, l) \in \Xi^1_j$, for all $k = -j, \dots, j$, $l = 1, 2, 3$.*

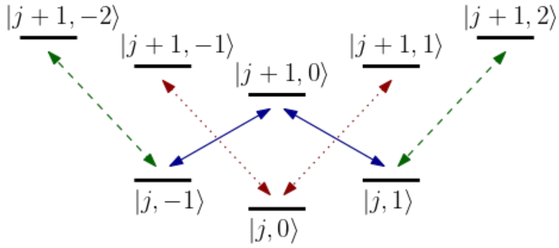


Figure 2: Graph of the transitions associated with the frequency λ_k^j between eigenstates $|j,k\rangle = |j,k,m\rangle := D^j_{k,m}$ (m fixed). Same-shaped arrows correspond to equal spectral gaps.

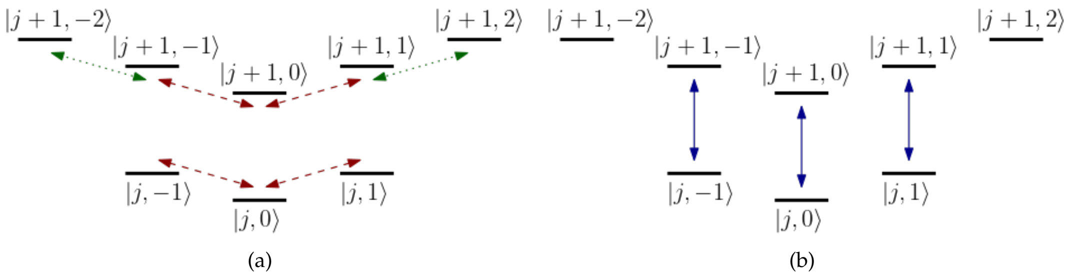


Figure 3: Transitions between states: (a) at frequency η_k; (b) at frequency σ^j. Same-shaped arrows correspond to equal spectral gaps.

Proof. Because of the selection rules (23), we only need to check if there are common spectral gaps in the spaces $\mathcal{M}_j$ and $\mathcal{M}_{j'}$ for $j' \neq j$.

We start by proving that $(\lambda_k^j, l), (\sigma^j, l) \in \Xi_j^0$ by showing that a spectral gap of the type λ_k^j (respectively, σ^j) is different from any spectral gap of the type $\lambda_{k'}^{j'}$, $\sigma^{j'}$, or $\eta_{k'}$ unless $\lambda_k^j = \lambda_{k'}^{j'}$ and $(k,j) = (k',j')$ (respectively, $\sigma^j = \sigma^{j'}$ and $j = j'$).

Using the explicit structure of the spectrum (20), any spectral gap of the type $\lambda_{k'}^{j'}$, $\sigma^{j'}$, or $\eta_{k'}$ can be written as

$$\Big|\frac{q_1}{I_2} + q_2\Big(\frac{1}{I_3} - \frac{1}{I_2}\Big)\Big|, \qquad q_1, q_2 \in \mathbb{Q}.$$

Since, moreover, $\frac{1}{I_2}$ and $\left(\frac{1}{I_3} - \frac{1}{I_2}\right)$ are $\mathbb{Q}$-linearly independent, one easily deduces that, indeed, $(\lambda_k^j, l), (\sigma^j, l) \in \Xi_j^0$.

Notice that the gaps of the type η_k correspond to internal pairings in the spaces $\mathcal{H}_j$. Henceforth, in order to prove that $(\eta_k, l) \in \Xi_j^1$ it is enough to check that η_k is different to any gap of the type $\lambda_{k'}^j, \sigma^j$. This fact has already been noticed in the proof of the first part of the statement. The proof of the lemma is then concluded. $\square$

Next, we introduce the family of excited modes at the frequencies λ_k^j,

$$\mathcal{F}_j := \{\mathcal{E}_{\lambda_k^j}(\mathrm{i}B_l), W_\mathrm{i}(\mathcal{E}_{\lambda_k^j}(\mathrm{i}B_l)) \mid l = 1,2,3,\ k = -j,\dots,j\},$$

where the operators $\mathcal{E}_\mu$ and W_ξ are defined in Section 2.1, and where, with a slight abuse of notation, we write B_l instead of $\Pi_{\mathcal{M}_j} B_l \Pi_{\mathcal{M}_j}$. Notice that $\mathcal{F}_j \subset \nu_j^0$ as it follows from Lemma 2, where ν_j^0 is defined as in (13).

Denote by $\mathrm{L}_j := \mathrm{Lie}(\mathcal{F}_j)$ the Lie algebra generated by the matrices in $\mathcal{F}_j$. Let us introduce the generalized Pauli matrices

$$E_{j,k} = e_{j,k} - e_{k,j}, \quad F_{j,k} = \mathrm{i}e_{j,k} + \mathrm{i}e_{k,j}, \quad D_{j,k} = \mathrm{i}e_{j,j} - \mathrm{i}e_{k,k},$$

where $e_{j,k}$ denotes the $(2j+1)^2 + (2(j+1)+1)^2$-square matrix whose entries are all zero, except the one at row j and column k, which is equal to 1. Consider again the lexicographic ordering $\rho : \{(l,k,m) \mid l = j, j+1, \ k,m = -l,\dots,l\} \to \mathbb{N}$. By a slight abuse of notation, also set $e_{(l,k,m),(l',k',m')} = e_{\rho(l,k,m),\rho(l',k',m')}$. The analogous identification can be used to define $E_{(l,k,m),(l',k',m')}, F_{(l,k,m),(l',k',m')}, D_{(l,k,m),(l',k',m')}$. The next proposition tells us how the elements in L_j look like. For a detailed proof, see Appendix A.

Proposition 1. *The matrices* $X_{(j,k,m),(j+1,k+1,m)} - X_{(j,-k,m),(j+1,-k-1,m)}$ *and* $X_{(j,k,m),(j+1,k+1,m\pm1)} + X_{(j,-k,m),(j+1,-k-1,m\pm1)}$ *are in* L_j *for all* $m = -j,\dots,j$ *and* $k = -j,\dots,j$, $k \neq 0$, *where* $X \in \{E, F\}$.

To break the degeneracy between k and $-k$ which appears in the elementary matrices that we found in Proposition 1, and obtain all the elementary matrices that one needs to generate $\mathfrak{su}(\mathcal{M}_j)$, we need to exploit the other two types of spectral gaps that we have introduced in (25) and (26) (see Figure 3).

Let us introduce the family of excited modes at the frequencies σ^j and η_k,

$$\mathcal{P}_j := \{\mathcal{E}_{\sigma^j}(\mathrm{i}B_l), W_{\mathrm{i}}(\mathcal{E}_{\sigma^j}(\mathrm{i}B_l)), \mathcal{E}_{\eta_k}(\mathrm{i}B_l), W_{\mathrm{i}}(\mathcal{E}_{\eta_k}(\mathrm{i}B_l)) \mid l = 1,2,3, \ k = -j,\dots,j\},$$

and notice that, by Lemma 2, $\mathcal{P}_j \subset \nu_j^1$ (cf. (13)). Therefore,

$$\widetilde{\mathcal{P}}_j := \{A, [B,C] \mid A, B \in \mathrm{L}_j, C \in \mathcal{P}_j\} \subset \mathcal{T}_j,$$

where we recall that $\mathcal{T}_j$ is the minimal ideal of $\mathrm{Lie}(\nu_j^1)$ containing ν_j^0.

The following proposition concludes the proof of Theorem 7.

Proposition 2. $\mathrm{Lie}(\widetilde{\mathcal{P}}_j) = \mathfrak{su}(\mathcal{M}_j)$.

For a proof of Proposition 2, see Appendix B. □

Remark 6. *The assumption $I_2/I_3 \notin \mathbb{Q}$ on the moments of inertia appearing in Theorem 7 is technical, and prevents the system from having both external resonances (as we saw in Lemma 2) and internal ones (as we will see in Lemma 3). Anyway, we have not proven that controllability fails if the ratio I_2/I_3 is rational.*

2.5 Reachable sets of the quantum genuine symmetric-top

In (22) we see that, when $\delta = (0,0,\delta_3)^t$, transitions $k \to k'$ are forbidden if $k \neq k'$. Thus, if the quantum system is prepared in the initial state $\psi(0)$ with $P_3\psi(0) = k\psi(0)$, the wave function ψ evolves in the subspaces $S_k = \overline{\mathrm{span}}\{D^j_{k,m} \mid j \in \mathbb{N}, m = -j,\dots,j\}$. The next theorem tells us that the restriction of (21) to this subspace is controllable.

Theorem 8. *Let $I_1 = I_2$ and fix $k \in \mathbb{Z}$. If $\delta = \begin{pmatrix} 0 \\ 0 \\ \delta_3 \end{pmatrix}$, $\delta_3 \neq 0$, then the Schrödinger equation (21) is an m-tracker in the Hilbert space S_k. In particular, $\mathrm{Reach}(\psi)$ is dense in $S_k \cap \mathcal{S}$ for all $\psi \in S_k \cap \mathcal{S}$.*

Proof. For every integer $j \geq |k|$, let $I_{j,k} := \{\rho(l,m) \mid l = j, j+1,\ m = -l,\dots,l\}$, where $\rho : \{(l,m) \mid l \geq |k|,\ m = -l,\dots,l\} \to \mathbb{N}$ is the lexicographic ordering. Then the graph $\mathcal{G}_k$ with vertices $\{I_{j,k}\}_{j=|k|}^{\infty}$ and edges $\{(I_{j,k}, I_{j',k}) \mid I_{j,k} \cap I_{j',k} \neq \emptyset\}$ is linear.

In order to apply Theorem 5 to the restriction of (21) to S_k, we should consider the projected dynamics onto $\mathcal{N}_{j,k} := \mathcal{L}_{j,k} \oplus \mathcal{L}_{j+1,k}$, where $\mathcal{L}_{l,k} := \mathrm{span}\{D^l_{k,m} \mid m = -l,\dots,l\}$. The only spectral gaps in S_k are $\sigma^j = |E^{j+1}_k - E^j_k| = \dfrac{j+1}{I_2}$, $j \geq |k|$. Notice that $(\sigma^j, l) \in \Xi^0_j$.

We write the electric potentials projected onto $\mathcal{N}_{j,k}$:

$$\begin{aligned}
\mathcal{E}_{\sigma^j}(\mathrm{i}B_1) &= \sum_{m=-j,\dots,j} a_{j,k,m}\delta_3 E_{(j,k,m),(j+1,k,m+1)} + a_{j,k,-m}\delta_3 E_{(j,k,m),(j+1,k,m-1)},\\
\mathcal{E}_{\sigma^j}(\mathrm{i}B_2) &= \sum_{m=-j,\dots,j} a_{j,k,m}\delta_3 F_{(j,k,m),(j+1,k,m+1)} - a_{j,k,-m}\delta_3 F_{(j,k,m),(j+1,k,m-1)},\\
\mathcal{E}_{\sigma^j}(\mathrm{i}B_3) &= \sum_{m=-j,\dots,j} -b_{j,k,m}\delta_3 F_{(j,k,m),(j+1,k,m)},
\end{aligned}$$

having used the explicit pairings (39), which can be found in Appendix B, and which describe the transitions excited by the frequency σ^j. Note that here the sum does not run over k since we are considering the dynamics restricted to S_k. We consider the family of excited modes

$$\mathcal{F}_{j,k} = \{\mathcal{E}_{\sigma^j}(\mathrm{i}B_l), W_{\mathrm{i}}(\mathcal{E}_{\sigma^j}(\mathrm{i}B_l)) \mid l = 1,2,3\} \subset \nu^0_j.$$

We claim that the Lie algebra generated by $\mathcal{F}_{j,k}$, seen as a subset of $\mathfrak{su}((2j+1)^2 + (2(j+1)+1)^2)$, is equal to $\mathfrak{su}((2j+1)^2 + (2(j+1)+1)^2)$. Such an identity has been proved in [8, Section 3.3], since the projection to $\mathcal{N}_{j,k}$ is isomorphic to an analogous projection for the linear molecule. Hence, we conclude that the system (21) is an m-tracker in S_k. $\square$

2.6 Non-controllability of the quantum orthogonal accidentally symmetric top

Let us consider separately the case where $\delta = (\delta_1, \delta_2, 0)^t$, left out by Theorem 7. The situation in which the dipole lies in the plane orthogonal to the symmetry axis of the molecule (that is, the *orthogonal* accidentally symmetric-top) is interesting from the point of view of chemistry, since the accidentally symmetric-top molecules present in nature are usually of that kind (see Figure 2.6). In order to study this problem, let us introduce the Wang functions

$$\begin{cases} S^j_{k,m,\gamma} := \dfrac{1}{\sqrt{2}}(D^j_{k,m} + (-1)^\gamma D^j_{-k,m}), & k = 1,\dots,j,\\ S^j_{0,m,0} = D^j_{0,m}, & k = 0, \end{cases}$$

for $j \in \mathbb{N}$, $m = -j,\dots,j$, and $\gamma = 0,1$. Due to the k-degeneracy $E^j_k = E^j_{-k}$ in the spectrum of the rotational Hamiltonian H, the functions $S^j_{k,m,\gamma}$ still form an orthogonal basis of eigenfunctions of H. Then we consider the change of basis $D^j_{k,m} \to e^{-\mathrm{i}k\theta} D^j_{k,m}$, and we choose $\theta \in [0, 2\pi)$ such that

$$\begin{cases} e^{-\mathrm{i}\theta}(\delta_2 + \mathrm{i}\delta_1) = \mathrm{i}\sqrt{\delta_1^2 + \delta_2^2},\\ e^{\mathrm{i}\theta}(\delta_2 - \mathrm{i}\delta_1) = -\mathrm{i}\sqrt{\delta_1^2 + \delta_2^2}. \end{cases} \tag{27}$$

System (27) describes a rotation in the complex plane of the vector $\delta_2 \pm \mathrm{i}\delta_1$, by the angle $\mp\theta$. The composition of this two changes of basis gives us the rotated Wang states $S^j_{k,m,\gamma}(\theta) := \dfrac{1}{\sqrt{2}}(e^{-\mathrm{i}k\theta} D^j_{k,m} + (-1)^\gamma e^{\mathrm{i}k\theta} D^j_{-k,m})$, for $k \neq 0$, and $S^j_{0,m,0} = D^j_{0,m}$.

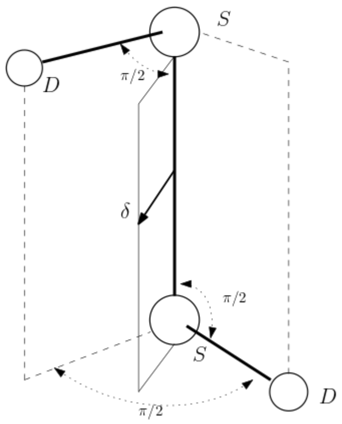


Figure 4: Diagram of the orthogonal accidentally symmetric molecule D_2S_2. The electric dipole δ lies in the orthogonal plane to the symmetry axis.

In the next theorem we express in this new basis a symmetry which prevents the system from being controllable.

Theorem 9. *Let* $I_1 = I_2$ *and* $\delta = \begin{pmatrix} \delta_1 \\ \delta_2 \\ 0 \end{pmatrix}$*, with* $\delta_1 \neq 0$ *or* $\delta_2 \neq 0$*. Then the parity of* $j + \gamma + k$ *is conserved.*

Proof. We need to prove that the pairings allowed by the controlled vector fields B_1, B_2 and B_3 conserve the parity of $j + \gamma + k$. To do so, let us compute

$$\begin{aligned} \langle S^j_{k,m,\gamma}(\theta), \mathrm{i}B_1 S^{j+1}_{k+1,m+1,\gamma}(\theta)\rangle &= -c_{j,k,m}e^{-\mathrm{i}\theta}(\delta_2 + \mathrm{i}\delta_1) + c_{j,k,m}e^{\mathrm{i}\theta}(\delta_2 - \mathrm{i}\delta_1) \\ &= -2\mathrm{i}c_{j,k,m}\sqrt{\delta_1^2 + \delta_2^2}, \\ \langle S^j_{k,m,\gamma}(\theta), \mathrm{i}B_1 S^{j+1}_{k+1,m+1,\gamma'}(\theta)\rangle &= -c_{j,k,m}e^{-\mathrm{i}\theta}(\delta_2 + \mathrm{i}\delta_1) - c_{j,k,m}e^{\mathrm{i}\theta}(\delta_2 - \mathrm{i}\delta_1) \\ &= 0, \qquad \gamma \neq \gamma', \end{aligned} \tag{28}$$

having used the expression of the Wang functions as linear combinations of Wigner functions, the explicit pairings (31) which can be found in Appendix A, and the choice we made of θ in (27). Then we also have

$$\begin{cases} \langle S^j_{k,m,\gamma}(\theta), \mathrm{i}B_1 S^j_{k+1,m+1,\gamma}(\theta)\rangle = 0, \\ \langle S^j_{k,m,\gamma}(\theta), \mathrm{i}B_1 S^j_{k+1,m+1,\gamma'}(\theta)\rangle = -2\mathrm{i}h_{j,k,m}\sqrt{\delta_1^2 + \delta_2^2}, \quad \gamma \neq \gamma', \end{cases} \tag{29}$$

having used this time the pairings (38), which can be found in Appendix B. From (28) and (29) we can see that the allowed transitions only depend on the parity of $j + \gamma$ and k, in fact we have either transitions between states of the form

$$\begin{cases} j + \gamma & \text{even} \\ k & \text{even} \end{cases} \longleftrightarrow \begin{cases} j' + \gamma' & \text{odd} \\ k' & \text{odd}, \end{cases}$$

or transitions between states of the form

$$\begin{cases} j+\gamma & \text{even} \\ k & \text{odd} \end{cases} \longleftrightarrow \begin{cases} j'+\gamma' & \text{odd} \\ k' & \text{even.} \end{cases}$$

The same happens if we replace $m+1$ with $m-1$ and $k+1$ with $k-1$ in (28) and (29). Because of the selection rules (23), these are the only transitions allowed by the field B_1. One can easily check, in the same way, that every transition induced by B_2, B_3 also conserves the parity of $j+\gamma+k$. $\square$

Appendix

A Proof of Proposition 1

In order to write down the matrices in $\mathcal{F}_j$, we need to study the resonances between the spectral gaps inside $\mathcal{M}_j$. First notice that the degeneracy $E_k^j = E_{-k}^j$ characterizes a symmetric-top molecule and we cannot remove it. We claim that there are no other internal resonances except those due to such a degeneracy. Indeed, we have already noticed in Lemma 2 that a spectral gap of the type λ_k^j (respectively, σ^j) is different from any spectral gap of the type $\lambda_{k'}^{j'}$, $\sigma^{j'}$, or $\eta_{k'}$ unless $\lambda_k^j = \lambda_{k'}^{j'}$ and $(k,j) = (k',j')$ (respectively, $\sigma^j = \sigma^{j'}$ and $j = j'$). Moreover, $\eta_k \neq \eta_{k'}$ if $k \neq k'$.

Hence we have the following.

Lemma 3 (Internal resonances). *Let $I_2/I_3 \notin \mathbb{Q}$. Then*

1. *λ_k^j-**resonances:** the equation*
$$|E_{k+1}^{j+1} - E_k^j| = |E_{s+h}^{j''} - E_s^{j'}|,\ j \le j' \le j'' \le j+1,\ -j' \le s \le j',\ h \in \{-1,0,1\},$$
implies that $j' = j$, $j'' = j+1$, $s = \pm k$, $s+h = \pm(k+1)$;
2. *η_k-**resonances:** the equation*
$$|E_{k+1}^{j} - E_k^j| = |E_{s+h}^{j''} - E_s^{j'}|,\ j \le j' \le j'' \le j+1,\ -j' \le s \le j',\ h \in \{-1,0,1\},$$
implies that $j' = j'' = j$ or $j' = j'' = j+1$ and $s = \pm k$, $s+h = \pm(k+1)$;
3. *σ^j-**resonances:** the equation*
$$|E_{k}^{j+1} - E_k^j| = |E_{s+h}^{j''} - E_s^{j'}|,\ j \le j' \le j'' \le j+1,\ -j' \le s \le j',\ h \in \{-1,0,1\},$$
implies that $j' = j$, $j'' = j+1$, $h = 0$, $s = \pm k$.

As a consequence of Lemma 3, part 1, if $I_2/I_3 \notin \mathbb{Q}$, the only transitions driven by the fields $\mathrm{i}B_l$, $l = 1,2,3$, excited at frequency λ_k^j, are the ones corresponding to the following matrix elements (written in the basis of $\mathcal{M}_j$ given by the Wigner functions) and can be computed using, e.g., [12, Table 2.1]:

$$\begin{cases} \langle D_{k,m}^j, \mathrm{i}B_1 D_{k+1,m\pm1}^{j+1}\rangle = -c_{j,k,\pm m}(\delta_2 + \mathrm{i}\delta_1), \\ \langle D_{k,m}^j, \mathrm{i}B_1 D_{k-1,m\pm1}^{j+1}\rangle = c_{j,-k,\pm m}(\delta_2 - \mathrm{i}\delta_1), \\ \langle D_{k,m}^j, \mathrm{i}B_2 D_{k+1,m\pm1}^{j+1}\rangle = \mp\mathrm{i}c_{j,k,\pm m}(\delta_2 + \mathrm{i}\delta_1), \\ \langle D_{k,m}^j, \mathrm{i}B_2 D_{k-1,m\pm1}^{j+1}\rangle = \pm\mathrm{i}c_{j,-k,\pm m}(\delta_2 - \mathrm{i}\delta_1), \\ \langle D_{k,m}^j, \mathrm{i}B_3 D_{k\pm1,m}^{j+1}\rangle = \pm\mathrm{i}d_{j,\pm k,m}(\delta_2 \pm \mathrm{i}\delta_1), \end{cases} \tag{30}$$

where

$$c_{j,k,m} := \frac{[(j+k+1)(j+k+2)]^{1/2}[(j+m+1)(j+m+2)]^{1/2}}{4(j+1)[(2j+1)(2j+3)]^{1/2}},$$

and

$$d_{j,k,m} := \frac{[(j+k+1)(j+k+2)]^{1/2}[(j+1)^2-m^2]^{1/2}}{2(j+1)[(2j+1)(2j+3)]^{1/2}}.$$

Now, using a symmetry agrument, we explain how to get rid of one electric dipole component between δ_1 and δ_2. Indeed, we shall explain in detail the meaning of the rotation (27) which we have already used in the proof of Theorem 9.

By the very definition of the Euler angles, one has that the rotation of angle θ around the symmetry axis a_3 is given by $\gamma \mapsto \gamma + \theta$. This rotation acts on the Wigner functions in the following way

$$D^j_{k,m}(\alpha,\beta,\gamma) \mapsto D^j_{k,m}(\alpha,\beta,\gamma+\theta) = e^{\mathrm{i}k\theta} D^j_{k,m}(\alpha,\beta,\gamma) =: D^j_{k,m}(\theta)(\alpha,\beta,\gamma),$$

having used the explicit expression of the symmetric states (19). Note that these rotated Wigner functions form again an orthogonal basis for $L^2(\mathrm{SO}(3))$ of eigenfunctions of the rotational Hamiltonian H, so we can also analyze the controllability problem in this new basis. In other words, the inertia ellipsoid of a symmetric-top molecule admits the circle S^1 as symmetry group. This is again a characterizing property of symmetric-top, which is no longer true for asymmetric-top molecules. In this new basis the matrix elements (corresponding to the frequency λ^j_k) of the controlled fields are

$$\begin{cases} \langle D^j_{k,m}(\theta), \mathrm{i}B_1 D^{j+1}_{k+1,m+1}(\theta)\rangle & = -c_{j,k,m} e^{-\mathrm{i}\theta}(\delta_2 + \mathrm{i}\delta_1), \\ \langle D^j_{k,m}(\theta), \mathrm{i}B_1 D^{j+1}_{k-1,m+1}(\theta)\rangle & = c_{j,-k,m} e^{\mathrm{i}\theta}(\delta_2 - \mathrm{i}\delta_1), \end{cases} \tag{31}$$

and the same happens for all the other transitions described in (30). So, the effect of this change of basis is that we are actually rotating the first two components of the dipole moment, by the angle θ. We can now choose $\theta \in [0, 2\pi)$ such that

$$e^{-\mathrm{i}\theta}(\delta_2 + \mathrm{i}\delta_1) = \sqrt{\delta_1^2 + \delta_2^2} = e^{\mathrm{i}\theta}(\delta_2 - \mathrm{i}\delta_1).$$

In other words, with this change of basis, we can assume without loss of generality that $\delta_1 = 0$, since we can rotate the vector $\delta_2 \pm \mathrm{i}\delta_1$ and get rid of its imaginary part (note that in (27) and in the proof of Theorem 9 we were rotating the vector $\delta_2 \pm \mathrm{i}\delta_1$ in the other sense, i.e., to get rid of its real part). This will simplify the expression of the controlled fields. Clearly, we could have alternatively chosen to set $\delta_2 = 0$.

Note that

$$\begin{aligned} W_{\mathrm{i}}(E_{(j,k,m),(j+1,k+1,n)}) &= \pm F_{(j,k,m),(j+1,k+1,n)}, \\ W_{\mathrm{i}}(F_{(j,k,m),(j+1,k+1,n)}) &= \mp E_{(j,k,m),(j+1,k+1,n)}, \end{aligned}$$

depending on whether I_2 is greater or smaller than I_3. We shall assume

$$\begin{aligned} W_{\mathrm{i}}(E_{(j,k,m),(j+1,k+1,n)}) &= -F_{(j,k,m),(j+1,k+1,n)}, \\ W_{\mathrm{i}}(F_{(j,k,m),(j+1,k+1,n)}) &= E_{(j,k,m),(j+1,k+1,n)}, \end{aligned}$$

since the computations in the two cases are completely analogous.

From the identity $[e_{j,k}, e_{n,m}] = \delta_{kn} e_{j,m} - \delta_{jm} e_{n,k}$ we get the useful bracket relations

$$[E_{j,k}, E_{k,n}] = E_{j,n}, \quad [F_{j,k}, F_{k,n}] = -E_{j,n}, \quad [E_{j,k}, F_{k,n}] = F_{j,n},$$

$$[E_{j,k}, F_{j,k}] = 2D_{j,k}, \quad [F_{j,k}, D_{j,k}] = 2E_{j,k}.$$

Moreover, two operators coupling no common states commute, that is,

$$[Y_{j,k}, Z_{j',k'}] = 0 \qquad \text{if } \{j,k\} \cap \{j',k'\} = \emptyset,$$

with $Y, Z \in \{E, F, D\}$.

Finally, we can conveniently represent the matrices corresponding to the controlled vector field (projected onto $\mathcal{M}_j$) in the rotated basis found with the symmetry argument. So, for each $k = -j, \ldots, j$, because of Lemma 3, part 1, and (30), we have

$$\begin{aligned}\mathcal{E}_{\lambda_k^j}(\mathrm{i}B_1) = \sum_{m=-j,\ldots,j} & -c_{j,k,m}\delta_2 E_{(j,k,m),(j+1,k+1,m+1)} - c_{j,k,-m}\delta_2 E_{(j,k,m),(j+1,k+1,m-1)} \\ & + c_{j,k,m}\delta_2 E_{(j,-k,m),(j+1,-k-1,m+1)} + c_{j,k,-m}\delta_2 E_{(j,-k,m),(j+1,-k-1,m-1)}, \end{aligned} \tag{32}$$

$$\begin{aligned}\mathcal{E}_{\lambda_k^j}(\mathrm{i}B_2) = \sum_{m=-j,\ldots,j} & -c_{j,k,m}\delta_2 F_{(j,k,m),(j+1,k+1,m+1)} + c_{j,k,-m}\delta_2 F_{(j,k,m),(j+1,k+1,m-1)} \\ & + c_{j,k,m}\delta_2 F_{(j,-k,m),(j+1,-k-1,m+1)} - c_{j,k,-m}\delta_2 F_{(j,-k,m),(j+1,-k-1,m-1)}, \end{aligned} \tag{33}$$

$$\mathcal{E}_{\lambda_k^j}(\mathrm{i}B_3) = \sum_{m=-j,\ldots,j} d_{j,k,m}\delta_2 F_{(j,k,m),(j+1,k+1,m)} - d_{j,k,m}\delta_2 F_{(j,-k,m),(j+1,-k-1,m)}, \tag{34}$$

where, with a slight abuse of notation, we write B_l instead of $\Pi_{\mathcal{M}_j} B_l \Pi_{\mathcal{M}_j}$.

Now we show how the sum over m in (32), (33) and (34) can be decomposed, in order to obtain that all the elementary matrices $X_{(j,k,m),(j+1,k+1,m\pm1)} + X_{(j,-k,m),(j+1,-k-1,m\pm1)}$ and $X_{(j,k,m),(j+1,k+1,m)} - X_{(j,-k,m),(j+1,-k-1,m)}$ are in L_j, for any $m, k = -j, \ldots, j$, where $X \in \{E, F\}$.

Let us first fix $k \neq 0$ and consider

$$\begin{aligned} & W_{\mathrm{i}}(\mathcal{E}_{\lambda_k^j}(\mathrm{i}B_3)) \\ & \qquad = \sum_{m=-j,\ldots,j} d_{j,k,m}\delta_2 E_{(j,k,m),(j+1,k+1,m)} - d_{j,k,m}\delta_2 E_{(j,-k,m),(j+1,-k-1,m)}, \end{aligned}$$

and the brackets

$$\begin{aligned}\mathrm{ad}^{2s}_{\mathcal{E}_{\lambda_k^j}(\mathrm{i}B_3)}(W_{\mathrm{i}}(\mathcal{E}_{\lambda_k^j}(\mathrm{i}B_3))) = \sum_{m=-j,\ldots,j} & (-1)^s 2^{2s} d_{j,k,m}^{2s+1}\delta_2^{2s+1} E_{(j,k,m),(j+1,k+1,m)} \\ & + (-1)^s 2^{2s}(-d_{j,k,m})^{2s+1}\delta_2^{2s+1} E_{(j,-k,m),(j+1,-k-1,m)}, \end{aligned}$$

for $s \in \mathbb{N}$, where $\mathrm{ad}_A(B) = [A, B]$ and $\mathrm{ad}_A^{n+1}(B) = [A, \mathrm{ad}_A^n(B)]$. Since $d_{j,k,m} = d_{j,k,-m}$, the invertibility of the Vandermonde matrix gives that

$$\begin{aligned} & E_{(j,k,m),(j+1,k+1,m)} + E_{(j,k,-m),(j+1,k+1,-m)} + \\ & - E_{(j,-k,m),(j+1,-k-1,m)} - E_{(j,-k,-m),(j+1,-k-1,-m)} \in \mathrm{L}_j, \end{aligned} \tag{35}$$

for $m = 0, \ldots, j$. In particular, $E_{(j,k,0),(j+1,k+1,0)} - E_{(j,-k,0),(j+1,-k-1,0)}$ is in L_j. Hence,

$$\begin{aligned} & \Big[\Big[\frac{\mathcal{E}_{\lambda_k}(\mathrm{i}B_1) - W_{\mathrm{i}}(\mathcal{E}_{\lambda_k}(\mathrm{i}B_2))}{2}, E_{(j,k,0),(j+1,k+1,0)} - E_{(j,-k,0),(j+1,-k-1,0)}\Big], \\ & E_{(j,k,0),(j+1,k+1,0)} - E_{(j,-k,0),(j+1,-k-1,0)}\Big] = -c_{j,k,0}\delta_2 E_{(j,k,0),(j+1,k+1,-1)} \\ & - c_{j,k,-1}\delta_2 E_{(j,k,1),(j+1,k+1,0)} - c_{j,k,0}\delta_2 E_{(j,-k,0),(j+1,-k-1,-1)} \\ & - c_{j,k,-1}\delta_2 E_{(j,-k,1),(j+1,-k-1,0)} \end{aligned} \tag{36}$$

is also in L_j. Define

$$\begin{aligned}Q_0 =& - c_{j,k,0}\delta_2 E_{(j,k,0),(j+1,k+1,-1)} - c_{j,k,-1}\delta_2 E_{(j,k,1),(j+1,k+1,0)}\\ &- c_{j,k,0}\delta_2 E_{(j,-k,0),(j+1,-k-1,-1)} - c_{j,k,-1}\delta_2 E_{(j,-k,1),(j+1,-k-1,0)},\\ Q_m =& - c_{j,k,-m}\delta_2 E_{(j,k,-m),(j+1,k+1,-m-1)} - c_{j,k,-m-1}\delta_2 E_{(j,k,m+1),(j+1,k+1,m)}\\ &- c_{j,k,-m}\delta_2 E_{(j,-k,-m),(j+1,-k-1,-m-1)} - c_{j,k,-m-1}\delta_2 E_{(j,-k,m+1),(j+1,-k-1,m)},\end{aligned}$$

if $0 < m < j$, and

$$Q_j = -c_{j,k,-j}\delta_2 E_{(j,k,-j),(j+1,k+1,-j-1)} - c_{j,k,-j}\delta_2 E_{(j,-k,-j),(j+1,-k-1,-j-1)}.$$

We have

$$\begin{aligned}&\Big[\Big[\sum_{m=s,\dots,j} Q_m, E_{(j,k,s),(j+1,k+1,s)} + E_{(j,k,-s),(j+1,k+1,-s)} - E_{(j,-k,s),(j+1,-k-1,s)}\\ &- E_{(j,-k,-s),(j+1,-k-1,-s)}\Big], E_{(j,k,s),(j+1,k+1,s)} + E_{(j,k,-s),(j+1,k+1,-s)}\\ &- E_{(j,-k,s),(j+1,-k-1,s)} - E_{(j,-k,-s),(j+1,-k-1,-s)}\Big] = Q_s,\end{aligned}$$

for $s = 1, \dots, j$. By iteration on s and because of (35), it follows that $Q_s \in \mathrm{L}_j$ for every $s = 0, \dots, j$. Now, since

$$\frac{Q_j}{-c_{j,k,-j}\delta_2} = E_{(j,k,-j),(j+1,k+1,-j-1)} + E_{(j,-k,-j),(j+1,-k-1,-j-1)} \in \mathrm{L}_j,$$

then

$$\begin{aligned}&\mathrm{ad}^2_{E_{(j,k,-j),(j+1,k+1,-j-1)}+E_{(j,-k,-j),(j+1,-k-1,-j-1)}}(E_{(j,k,j),(j+1,k+1,j)}\\ &+ E_{(j,k,-j),(j+1,k+1,-j)} - E_{(j,-k,j),(j+1,-k-1,j)} - E_{(j,-k,-j),(j+1,-k-1,-j)})\\ &= E_{(j,k,-j),(j+1,k+1,-j)} - E_{(j,-k,-j),(j+1,-k-1,-j)} \in \mathrm{L}_j,\end{aligned}$$

which, in turns, implies that

$$\begin{aligned}&\mathrm{ad}^2_{E_{(j,k,-j),(j+1,k+1,-j)}-E_{(j,-k,-j),(j+1,-k-1,-j)}}(Q_{j-1})\\ &= -c_{j,k,-j+1}E_{(j,k,-j+1),(j+1,k+1,-j)} - c_{j,k,-j+1}E_{(j,-k,-j+1),(j+1,-k-1,-j)} \in \mathrm{L}_j.\end{aligned}$$

Iterating the argument,

$$E_{(j,k,m),(j+1,k+1,m)} - E_{(j,-k,m),(j+1,-k-1,m)} \in \mathrm{L}_j, \quad m = -j, \dots, j \tag{37}$$

and $E_{(j,k,m),(j+1,k+1,m-1)} + E_{(j,-k,m),(j+1,-k-1,m-1)}$ are in L_j for $m = -j, \dots, j$.

By the same argument as above, with $\dfrac{\mathcal{E}_{\lambda^j_k}(\mathrm{i}B_1) - W_\mathrm{i}(\mathcal{E}_{\lambda^j_k}(\mathrm{i}B_2))}{2}$ replaced by

$$\begin{aligned}\frac{\mathcal{E}_{\lambda^j_k}(\mathrm{i}B_1) + W_\mathrm{i}(\mathcal{E}_{\lambda^j_k}(\mathrm{i}B_2))}{2} = &\sum_{m=-j,\dots,j} -c_{j,k,m}E_{(j,k,m),(j+1,k+1,m+1)}\\ &+ c_{j,k,m}E_{(j,-k,m),(j+1,-k-1,m+1)}\end{aligned}$$

in (36) we also have that $E_{(j,k,m),(j+1,k+1,m+1)} + E_{(j,-k,m),(j+1,-k-1,m+1)}$ is in L_j for all $m = -j, \dots, j$.

If we now replace $\dfrac{\mathcal{E}_{\lambda_k^j}(\mathrm{i}B_1) - W_{\mathrm{i}}(\mathcal{E}_{\lambda_k^j}(\mathrm{i}B_2))}{2}$ with

$$\frac{\mathcal{E}_{\lambda_k^j}(\mathrm{i}B_2) + W_{\mathrm{i}}(\mathcal{E}_{\lambda_k^j}(\mathrm{i}B_1))}{2} = \sum_{m=-j,\dots,j} -c_{j,k,-m}F_{(j,k,m),(j+1,k+1,m-1)} + c_{j,k,-m}F_{(j,-k,m),(j+1,-k-1,m-1)}$$

or

$$\frac{\mathcal{E}_{\lambda_k^j}(\mathrm{i}B_2) - W_{\mathrm{i}}(\mathcal{E}_{\lambda_k^j}(\mathrm{i}B_1))}{2} = \sum_{m=-j,\dots,j} -c_{j,k,m}F_{(j,k,m),(j+1,k+1,m+1)} + c_{j,k,m}F_{(j,-k,m),(j+1,-k-1,m+1)},$$

the arguments above prove that both $F_{(j,k,m),(j+1,k+1,m)} - F_{(j,-k,m),(j+1,-k-1,m)}$ and $F_{(j,k,m),(j+1,k+1,m\pm1)} + F_{(j,-k,m),(j+1,-k-1,m\pm1)}$ are in L_j for all $m = -j, \dots, j$.

B Proof of Proposition 2

Using again [12, Table 2.1] we write the pairings

$$\begin{cases} \langle D^j_{k,m}, \mathrm{i}B_1 D^j_{k+1,m\pm1}\rangle = \mp h_{j,k,\pm m}(\delta_2 + \mathrm{i}\delta_1), \\ \langle D^j_{k,m}, \mathrm{i}B_1 D^j_{k-1,m\pm1}\rangle = \mp h_{j,-k,\pm m}(\delta_2 - \mathrm{i}\delta_1), \\ \langle D^j_{k,m}, \mathrm{i}B_2 D^j_{k+1,m\pm1}\rangle = -\mathrm{i}h_{j,k,\pm m}(\delta_2 + \mathrm{i}\delta_1), \\ \langle D^j_{k,m}, \mathrm{i}B_2 D^j_{k-1,m\pm1}\rangle = -\mathrm{i}h_{j,-k,\pm m}(\delta_2 - \mathrm{i}\delta_1), \\ \langle D^j_{k,m}, \mathrm{i}B_3 D^j_{k\pm1,m}\rangle = -\mathrm{i}q_{j,\pm k,m}(\delta_2 \pm \mathrm{i}\delta_1), \end{cases} \tag{38}$$

where

$$h_{j,k,m} := \frac{[j(j+1) - k(k+1)]^{1/2}[j(j+1) - m(m+1)]^{1/2}}{4j(j+1)},$$
$$q_{j,k,m} := \frac{[j(j+1) - k(k+1)]^{1/2}m}{2j(j+1)}.$$

Moreover,

$$\begin{cases} \langle D^j_{k,m}, \mathrm{i}B_1 D^{j+1}_{k,m\pm1}\rangle =: a_{j,k,\pm m}\delta_3, \\ \langle D^j_{k,m}, \mathrm{i}B_2 D^{j+1}_{k,m\pm1}\rangle = \pm\mathrm{i}a_{j,k,\pm m}\delta_3, \\ \langle D^j_{k,m}, \mathrm{i}B_3 D^{j+1}_{k,m}\rangle =: -\mathrm{i}b_{j,k,m}\delta_3, \end{cases} \tag{39}$$

where

$$a_{j,k,m} := \frac{[(j+1)^2 - k^2]^{1/2}[(j+m+1)(j+m+2)]^{1/2}}{2(j+1)[(2j+1)(2j+3)]^{1/2}},$$
$$b_{j,k,m} := \frac{[(j+1)^2 - k^2]^{1/2}[(j+1)^2 - m^2]^{1/2}}{(j+1)[(2j+1)(2j+3)]^{1/2}}.$$

Note that the $k \to k$ transitions are driven by δ_3. Since we are still assuming that $\delta_1 = 0$, and because of Lemma 3, parts 2 and 3, the expression of the controlled fields excited at the frequencies η_k and σ^j are

$$\mathcal{E}_{\eta_k}(\mathrm{i}B_1) = \sum_{\substack{l=j,j+1,\\ m=-l,\dots,l-1}} -h_{l,k,m}\delta_2 E_{(l,k,m),(l,k+1,m+1)} - h_{l,k,m}\delta_2 E_{(l,-k,m),(l,-k-1,m+1)}$$
$$\sum_{\substack{l=j,j+1,\\ m=-l+1,\dots,l}} h_{l,k,-m}\delta_2 E_{(l,k,m),(l,k+1,m-1)} + h_{l,k,-m}\delta_2 E_{(l,-k,m),(l,-k-1,m-1)}, \tag{40}$$

$$\mathcal{E}_{\eta_k}(\mathrm{i}B_2) = \sum_{\substack{l=j,j+1,\\ m=-l,\dots,l-1}} -h_{l,k,m}\delta_2 F_{(l,k,m),(l,k+1,m+1)} - h_{l,k,m}\delta_2 F_{(l,-k,m),(l,-k-1,m+1)}$$
$$\sum_{\substack{l=j,j+1,\\ m=-l+1,\dots,l}} -h_{l,k,-m}\delta_2 F_{(l,k,m),(l,k+1,m-1)} - h_{l,k,-m}\delta_2 F_{(l,-k,m),(l,-k-1,m-1)}, \tag{41}$$

$$\mathcal{E}_{\eta_k}(\mathrm{i}B_3) = \sum_{\substack{l=j,j+1,\\ m=-l,\dots,l}} -q_{l,k,m}\delta_2 F_{(l,k,m),(l,k+1,m)} - q_{l,k,m}\delta_2 F_{(l,-k,m),(l,-k-1,m)}, \tag{42}$$

and

$$\mathcal{E}_{\sigma^j}(\mathrm{i}B_1) = \sum_{m,k=-j,\dots,j} a_{j,k,m}\delta_3 E_{(j,k,m),(j+1,k,m+1)} + a_{j,k,-m}\delta_3 E_{(j,k,m),(j+1,k,m-1)}, \tag{43}$$
$$\mathcal{E}_{\sigma^j}(\mathrm{i}B_2) = \sum_{m,k=-j,\dots,j} a_{j,k,m}\delta_3 F_{(j,k,m),(j+1,k,m+1)} - a_{j,k,-m}\delta_3 F_{(j,k,m),(j+1,k,m-1)},$$
$$\mathcal{E}_{\sigma^j}(\mathrm{i}B_3) = \sum_{m,k=-j,\dots,j} -b_{j,k,m}\delta_3 F_{(j,k,m),(j+1,k,m)}.$$

Note that in $\mathcal{E}_{\eta_k}(-iB_3)$ the term for $m = 0$ vanishes, since $q_{j,k,0} = 0$ for every j, k.

To decouple all the m-degeneracies in the excited modes, we just make double brackets with the elementary matrices that we have obtained above. As an example, using (37) we can decouple the $m \to m$ transitions corresponding to the frequency σ^j by considering

$$\begin{aligned}&[[W_{\mathrm{i}}(\mathcal{E}_{\sigma^j}(\mathrm{i}B_3)), E_{(j,k,m),(j+1,k+1,m)} - E_{(j,-k,m),(j+1,-k-1,m)}],\\ &E_{(j,k,m),(j+1,k+1,m)} - E_{(j,-k,m),(j+1,-k-1,m)}]\\ &= E_{(j,k,m),(j+1,k,m)} - E_{(j,-k,m),(j+1,-k,m)} \in \mathrm{Lie}(\widetilde{\mathcal{P}}_j).\end{aligned}$$

Making every possible double brackets as above, we obtain, for $X \in \{E, F\}$, that

$$X_{(j,k,m),(j+1,k,m\pm h)} + X_{(j,-k,m),(j+1,-k,m\pm h)} \in \mathrm{Lie}(\widetilde{\mathcal{P}}_j), \quad k \neq 0, \tag{44}$$

when we start from the matrices in (43), and that

$$X_{(l,k,m),(l,k+1,m)} + X_{(l,-k,m),(l,-k-1,m)}, X_{(l,k,m),(l,k+1,m\pm1)} + X_{(l,-k,m),(j,-k-1,m\pm1)}$$

are in $\mathrm{Lie}(\widetilde{\mathcal{P}}_j)$, $l = j, j+1$, $m, k \neq 0$, when we start from the matrices in (40), (41), (42). Now we can also generate the missing $k = 0$ elements of (37) by making double brackets with $X_{(j+1,1,m),(j+1,2,m)} + X_{(j+1,-1,m),(j+1,-2,m)} \in \mathrm{Lie}(\widetilde{\mathcal{P}}_j)$. As an example, we have that

$$\begin{aligned}&[[\mathcal{E}_{\lambda_0^j}(\mathrm{i}B_3), F_{(j+1,1,m),(j+1,2,m)} + F_{(j+1,-1,m),(j+1,-2,m)}],\\ &F_{(j+1,1,m),(j+1,2,m)} + F_{(j+1,-1,m),(j+1,-2,m)}]\\ &= F_{(j,0,m),(j+1,1,m)} + F_{(j,0,m),(j+1,-1,m)} \in \mathrm{Lie}(\widetilde{\mathcal{P}}_j).\end{aligned}$$

Moreover, also the $m = 0$ elements in the transitions (42) are in $\mathrm{Lie}(\widetilde{\mathcal{P}}_j)$, as one can check by making a bracket between two transitions obtained in (37) and (44). For example,

$$[E_{(j,k,0),(j+1,k+1,0)} - E_{(j,-k,0),(j+1,-k-1,0)}, E_{(j+1,k+1,0),(j,k+1,0)} \\ + E_{(j+1,-k-1,0),(j,-k-1,0)}] = E_{(j,k,0),(j,k+1,0)} - E_{(j,-k,0),(j,-k-1,0)} \in \mathrm{Lie}(\widetilde{\mathcal{P}}_j).$$

Finally, we apply a three-waves mixing argument (Figure B) in order to decouple the sum over k and $-k$ in every elementary matrices: consider the bracket between the following elements in $\mathrm{Lie}(\widetilde{\mathcal{P}}_j)$

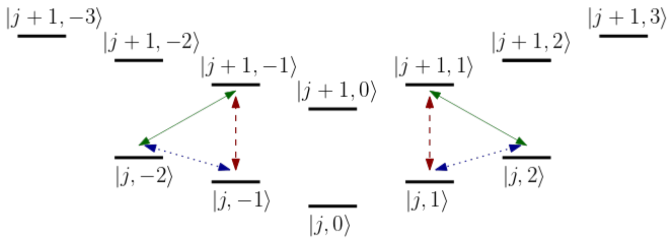


Figure 5: Three-waves mixing around $k = 1, -1$. The same-shaped arrows correspond to equal spectral gaps, and thus, coupled transitions. The goal of the three-waves mixing is to decouple those arrows.

$$[E_{(j,k+1,m),(j,k,m)} + E_{(j,-k-1,m),(j,-k,m)}, E_{(j,k,m),(j+1,k,m)} + E_{(j,-k,m),(j+1,-k,m)}] \\ = E_{(j,k+1,m),(j+1,k,m)} + E_{(j,-k-1,m),(j+1,-k,m)} \in \mathrm{Lie}(\widetilde{\mathcal{P}}_j), \quad k \neq 0,$$

and notice that from (37) we already have that

$$E_{(j,k+1,m),(j+1,k,m)} - E_{(j,-k-1,m),(j+1,-k,m)}$$

is in $\mathrm{Lie}(\widetilde{\mathcal{P}}_j)$, and hence $E_{(j,k+1,m),(j+1,k,m)}$ and $E_{(j,-k-1,m),(j+1,-k,m)}$ are in $\mathrm{Lie}(\widetilde{\mathcal{P}}_j)$. In this way we can break every k-degeneracy, and finally obtain that $\mathrm{Lie}(\widetilde{\mathcal{P}}_j) = \mathfrak{su}(\mathcal{M}_j)$, which concludes the proof.

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